



Concentrations and Sources of Aliphatic and Aromatic Hydrocarbons in Babolsar Coastal Sediments in the Caspian Sea

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ABSTRACT

This paper presents concentrations and sources of Aliphatic and Aromatic Hydrocarbons in the sediments from Babolsar coastal area and the inlet of Babolrood River in the southern side of the Caspian Sea. The concentration of hydrocarbons in 13 sediment samples from the study area were measured by gas chromatography (GC). Total Petroleum Hydrocarbon (TPH) concentrations in sediment samples in the coastal area ranged from 115 to 201 $\mu\text{g/g}$. In the inlet samples, TPH concentrations were close to each other and ranged from 294 to 367 $\mu\text{g/g}$. The TPH results showed moderate level of oil pollution in the study area. Total Polycyclic Aromatic Hydrocarbons (ΣPAHs) concentrations in sediment samples inside the inlet ranged from 498 to 702 ng/g , indicating moderate level of pollution. Concentrations of ΣPAHs in sediment samples in the coastal area ranged from 341 to 1703 ng/g , indicating moderate to less than significant level of pollution. Developed indices for pollutant origins showed that hydrocarbons in all sediment samples collected in the study area had petrogenic origin. The results also showed the Babolrood River as the main source of oil pollution in the sediments in the study area.

KEYWORDS: Caspian Sea, Babolrood River, Sediment, Oil pollution, TPH, PAH, Petroleum Hydrocarbon

INTRODUCTION

The Caspian Sea is the largest inland body of water on Earth. Its unique marine environment is the most important sturgeon habitat in the world and its fishery resources and huge oil and natural gas reserves have made it a major source of income for its littoral countries (USGS, 2010; Koutsouradi et al., 2018; Ruban and Khodorevskaya, 2011; Korshenko and Gul, 2005). However, the marine environment of the Caspian Sea due to extensive human exploitation and discharge of large magnitudes of urban, industrial and agricultural waste is under extensive pressure (Lattuada et al., 2019; Ranjbar and Zaker, 2016; Kaplin, 1995, Kosarev and Yablonskaya, 1994). In general, for all kind of pollution studies in the Caspian Sea, there were marked small-scale patches in time and space of very high concentration (Korshenko and Gul, 2005). The Caspian Sea is exposed to significant petroleum pollution mainly caused by offshore oil production and tanker traffic and accidents (Lattuada et al., 2019; Abilov et al., 1999; Tolosa et al., 2004; Korshenko and Gul, 2005).

The occurrence of any oil spill in the coastal area is associated with damage to the marine environment. Depending on the time, place, toxicity, and concentration of the oil spill,

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exposure to oil spill can lead to the death of various organisms and could cause harmful effects such as reduced reproduction, inappropriate growth, difficulty in feeding mechanisms, and diminished defense capacity against diseases (Clark, 2001; Law and Klungsoyr, 2000). The study of concentration and source of hydrocarbons in sediments is a common and effective way for understanding the level of oil pollution in coastal environment (Tolosa et al., 2004, 2005, 2009, Mirvakili and Zaker, 2014, Melé et al., 2018).

Aliphatic and aromatic hydrocarbons in coastal sediments could have petrogenic origin or could be the result of natural inputs from terrestrial plant waxes, marine phytoplankton, volcanic eruptions, biomass combustion and natural oil seeps (Tolosa et al., 2004; Meyers, 2003). Aliphatic hydrocarbons include straight chain alkanes, branched chain alkanes, cyclo alkanes and isoprenoids compounds, including biomarkers (Clark, 2001; Tolosa et al., 2004). Biomarkers can be used to provide information on the source of hydrocarbons and the extent of weathering of the oil spill in coastal sediments (Pu et al., 2017; Wang et al., 2014; Peng et al., 2016; Tarasov et al., 2013). Some polycyclic aromatic hydrocarbons (PAHs) are known to be highly pollutant and potent carcinogens with destructive effects on human health (Clark and Finley, 1973; Abdel-Shafy and Mansour, 2016). PAHs in coastal sediments could have several origins including incomplete combustion of fossil fuels, discharge of petroleum and its products, and biological processes. (Tolosa et al., 2004; Lourenço et al., 2013). Tolosa et al. (2004) conducted the first extensive study on the spatial distribution and sources of aliphatic and PAHs in coastal sediments from the Caspian Sea. Their results showed moderate to high petrogenic contamination in sediments from South Caspian Sea; high levels in particular were found in the offshore oil fields near Baku, Azerbaijan. Contaminant patterns indicated PAHs were mainly from fossil sources. Natural PAHs from in situ biological activity were found only important at a few sites influenced by the Iranian river discharges.

Zaker et al. (2012) studied the concentration and source of petroleum hydrocarbons in the coastal sediments of Bandar Anzali in south-western coast of the Caspian Sea. TPH concentrations in the surface sediments indicated low to medium level of petroleum pollution. The Concentrations of Σ PAHs in the sediments indicated absence of contamination in the sediment samples.

Azimi et al. (2014) studied the PAH levels in surface sediments of the Anzali Wetland and Caspian Sea. Their results showed PAH concentrations were higher in the coastal area of Caspian Sea. In both Anzali Wetland and the Caspian Sea coast, PAHs had petrogenic sources. The study indicated the toxicity for benthic organisms was low to medium.

Nemati et al. (2015), studies the origin and level of PAHs contamination in the sediments from the coasts of the Guilan Province in southwest Caspian Sea. The results showed petrogenic source for PAHs and indicated that total PAH concentrations at all sites were below the effects range-low.

Eghtesadi et al. (2014) studies the level and source of PAHs in the surface sediments from the Gorgan Bay, in the southeast coast of the Caspian Sea. Their results showed PAHs came from mixed and pyrogenic origin.

In this paper, using lab analysis, source of hydrocarbons and oil pollution level in sediment samples from Babolsar coastal area in the southern coast of the Caspian Sea are studied (Figs. 1 and 2). The study area is located at 36° 43.25'N and 52°, 39.50' E in Iran. It covered a 1100m-long part of the end of the Babolrood River and a band of coastal waters in the east of the river mouth which extended 1300m along-shore and 1700m cross-shore (Figs. 1 and 2).

The inlet of Babolrood River, during the past 30 years, has been used as a fishery port for a large number of Kilka fishing vessels. The waste oil from the vessels together with the

discharge of runoff and wastewater from cities of Babol and Babolsar into the river are main sources of oil pollution in the study area.



Figure 1. Study area

MATERIALS AND METHODS

Surface seabed sediment samples were collected at 13 stations in Babolrood River inlet and Babolsar coastal waters in the southern coast of the Caspian Sea in the summer of 2014 (Figs. 1 and 2). In the southern coastal waters of the Caspian Sea, the prevailing currents are mainly from west to east (Zaker et al., 2011), and consequently, the river plume transports mainly towards eastern side of the river mouth. Therefore, considering the river plume as the main source of pollution in the study area, the location of sediment sampling stations 1 to 9 were selected in the eastern side of the river mouth in the area most affected by the plume. Stations 10, 12 and 13 were located inside the river inlet. Station 11, as a control point, was located on the western side of the river mouth where sediments were least affected by the river plume (Fig. 2). Geographical coordinates and water depth of the sampling stations are presented in Table 1.

The UNEP/IOC/IAEA standard method (No. 20) (UNEP, 1992) was employed for sediment sampling. The sediments were sampled using a standard Van Veen Grab Sampler with 25x25 cm cross section. About 200 to 300 grams of wet sediment was taken from the top 5 centimeter of surface seabed sediment. The collected samples were packed and transferred to laboratory using the USEPA-sw-846 standard method. The standard method of American Association of Environmental Protection (USEPA-SW-846#3540C) named SOXHLET was used for the preparation of samples and extraction of petroleum hydrocarbons from them. The samples were passed through a 63 microns sieve (<63) before the lab analysis. A Gas chromatography device (GC-FID) Model VARRIAN was used for determining the concentration of Total Petroleum Hydrocarbons (TPHs) and Aliphatic and Aromatic compounds in the sediment samples. In addition, the concentration of normal alkanes (n-C12 - n-C30) and abundance of PAH compounds were measured. Quality assurance and quality control (QA/QC) for PAHs analysis were conducted according to the method of Tolosa et al., (2005).

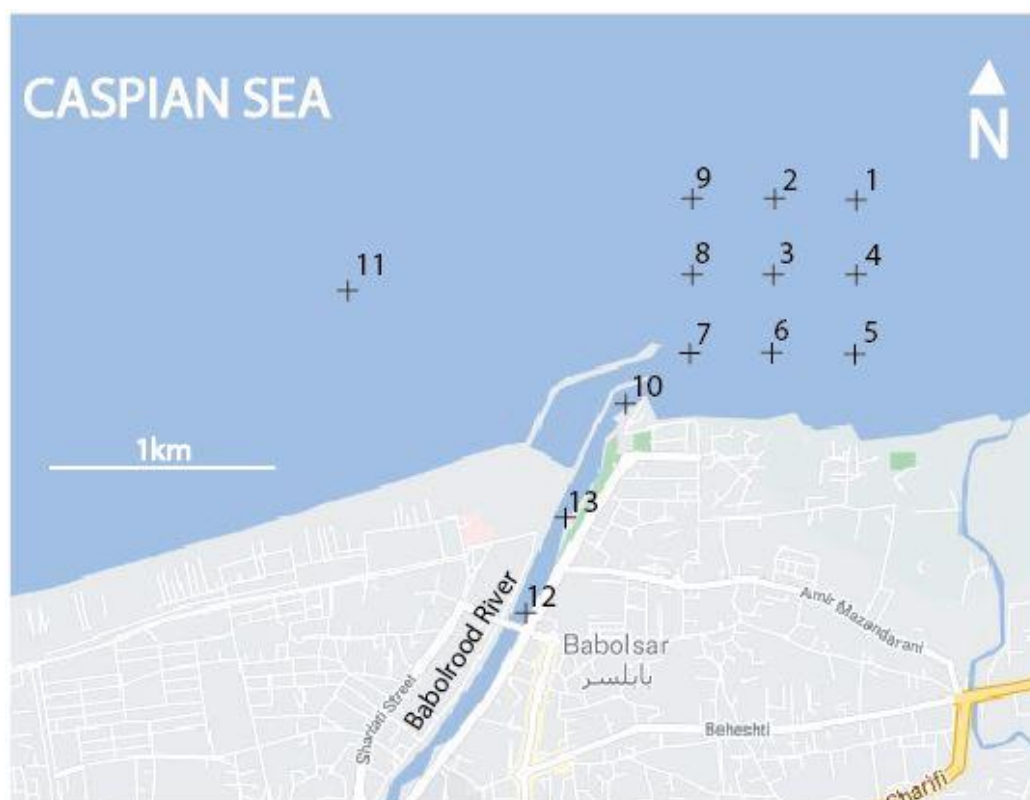


Figure 2. Location of sediment sampling stations

Table 1. Location and depth of sediment sampling stations

Station	Depth (m)	Latitude	Longitude	Sediment Type
1	4.5	36° 43.58' N	52° 40.17' E	fine sand
2	4.5	36° 43.58' N	52° 39.83' E	fine sand
3	3.5	36° 43.31' N	52° 39.83' E	fine sand
4	3.5	36° 43.31' N	52° 40.17' E	fine sand
5	1.5	36° 43.04' N	52° 40.17' E	fine sand
6	2.5	36° 43.04' N	52° 39.83' E	fine sand
7	3.0	36° 43.04' N	52° 39.52' E	fine sand
8	4.0	36° 43.32' N	52° 39.53' E	fine sand
9	4.5	36° 43.59' N	52° 39.53' E	fine sand
10	2.0	36° 42.91' N	52° 39.30' E	fine silty sand
11	3.5	36° 43.32' N	52° 38.26' E	fine sand
12	1.5	36° 42.40' N	52° 38.99' E	fine silty sand
13	2.5	36° 42.65' N	52° 39.14' E	fine silty sand

RESULTS AND DISCUSSIONS

Grain size distribution results, using standard techniques, showed fine sand and fine silty sand types sediments for coastal and river sampling stations, respectively (Table 1).

TPH concentrations in the sediment samples ranged from 53 $\mu\text{g/g}$ to 367 $\mu\text{g/g}$ (Fig. 3). The highest amounts were found in the river inlet at stations 10, 12 and 13 (Fig. 2), with values 294, 367 and 360 $\mu\text{g/g}$, respectively (Fig. 3). The lowest amount was observed at coastal station 11 where exposure to the river plume was minimum. TPH concentrations at coastal stations 1 to 9 (Fig. 2) ranged from 115 to 201 $\mu\text{g/g}$ (Fig. 3).

TPH concentrations decreased towards offshore and had amounts close to each other in the samples with similar distances from the coastline. Satellite images (not shown here) showed

stations 5, 6 and 7 were most affected by the river plume. In these stations, from the river mouth towards east, TPH concentration decreased significantly. The above results showed that the Babolrood River was the main source for petroleum hydrocarbons in the sediments from the study area.

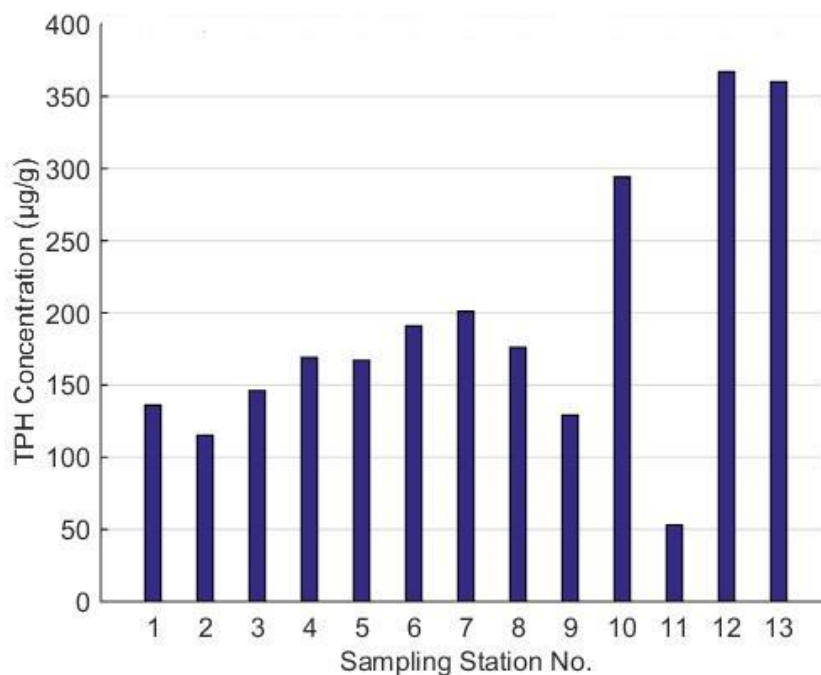


Figure 3. TPH concentration in sediment samples.

Commendatore and Esteves (2007) considered three TPH concentration levels for oil pollution in coastal sediments: low ($< 10 \mu\text{g/g}$), low to moderate ($10\text{-}100 \mu\text{g/g}$) and moderate to high ($100\text{-}1000 \mu\text{g/g}$). Readman et al. (2002) considered the sediments with TPH concentrations above $100 \mu\text{g/g}$ as polluted. Tolosa et al. (2004) considered TPH concentrations higher than $500 \mu\text{g/g}$ indicative of significant pollution and values below $10 \mu\text{g/g}$ as unpolluted sediments. Considering the above criteria, the level of oil pollution at station 11 was low and in all other stations were moderate to high.

Among the 100 polycyclic aromatic compounds identified so far, the US Environmental Protection Agency (EPA) has introduced 16 PAH compounds with carcinogenic and mutagenic effects on humans and animals as the leading pollutants (Tolosa et al., 2005; Bengner et al., 2010; Gao et al., 2018). These PAHs are Naphthalene, Acenaphthylene, Acenaphthene, Fluorene, Phenanthrene, Anthracene, Fluoranthene, Pyrene, Benzo[a]anthracene, Chrysene, Benzo[b]fluoranthene, Benzo[k]fluoranthene, Benzo[a]pyrene, Dibenzo[ah]anthracene, Indeno[123-cd]pyrene and Benzo[ghi]perylene. In this study, the concentrations of the above PAH compounds in the sediment samples were measured and the results are presented in Table 2.

The concentrations of the PAH compounds ranged from <10 to 579 ng/g . Fossil PAHs of Naphthalene, Acenaphthylene, Fluorene, Phenanthrene and Fluoranthene were observed at all stations and had higher concentrations than other compounds, indicating oil as the main source of PAHs in the study area (Tolosa et al., 2004). Indeno[123-cd]pyrene, Benzo[ghi]perylene and Dibenzo[ah]anthracene concentrations were very small at all station. Benzo[a]pyrene was observed at only two stations.

Table 2. Concentration of the main PAH compounds in the sediment samples (ng/g)

PAH (ng/g)	Stations												
	1	2	3	4	5	6	7	8	9	10	11	12	13
Naphthalene	180	181	160	247	333	190	270	156	148	323	48	342	259
Acenaphthylene	119	107	31	24	34	24	237	25	24	44	12	34	32
Fluorene	190	291	284	80	159	56	95	76	57	130	23	112	81
Phenanthrene	125	457	579	108	543	67	191	108	53	111	*	138	71
Fluoranthene	231	196	129	32	181	17	199	98	12	11	*	14	*
Pyrene	72	69	368	65	210	40	170	125	31	24	*	42	29
Acenaphthene	34	17	*	20	14	15	100	19	16	11	15	20	16
Anthracene	51	90	26	*	39	*	327	*	*	*	*	*	*
Benzo[b]fluoranthene	76	79	34	*	53	*	31	*	*	*	*	*	*
Benzo[k]fluoranthene	78	81	35	*	54	*	*	*	*	*	*	*	*
Chrysene	11	14	14	*	18	*	42	*	*	*	*	*	*
Benzo[a]anthracene	31	26	*	*	18	*	41	*	*	*	*	*	*
Benzo[a]pyrene	21	12	*	*	*	*	*	*	*	*	*	*	*
Indeno[123-cd]pyrene	*	*	*	*	*	*	*	*	*	*	*	*	*
Benzo[ghi]perylene	*	*	*	*	*	*	*	*	*	*	*	*	*
Dibenzo[ah]anthracene	*	*	*	*	*	*	*	*	*	*	*	*	*
Σ PAHs (ng/g)	1219	1620	1660	576	1656	409	1703	607	341	654	98	702	498

* <10 ng/g

Σ PAHs concentrations ranged from 98 to 1703 ng/g. Inside the river at stations 10, 12 and 13, Σ PAHs concentrations were 654, 702 and 498 ng/g, respectively and close to each other. In the coastal area, the minimum concentration of Σ PAHs with the amount of 98 ng/g was observed at station 11 where the river plume effect was insignificant. At the other coastal stations, the Σ PAHs concentrations ranged from a minimum of 341 ng/g at station 9, located further offshore, to a maximum of 1703 ng/g at station 7 near the river mouth. No specific trend was observed in the distribution of Σ PAHs concentrations in the coastal area.

PAHs concentrations were all less than the NOAA (1999) Sediment Quality Guideline value for the effects range low of 4000 ng/g dry weight, representing a "Minimal-effects" range with condition in which effects rarely be observed (Long et al., 1995). Baumard et al. (1998) considered 2 levels for Σ PAHs concentration in the sediments: low to moderately polluted (100-1000 ng/g) and highly polluted (>5000 ng/g). Considering these levels, sediment samples at all station, except at station 11 with no pollution, were moderately polluted.

The origin of PAHs in the sediments was investigated using Phen/An and Chr/BaA indexes. The Phen/An index uses the ratio of the amount of Phenanthrene compounds to the amount of Anthracene compounds for distinguishing the source of PAHs in marine sediments (Neff et al., 2005). The amount of Phen/An index for petroleum is high and has been reported to be about 60 (Yim et al., 2007). Chr/BaA index uses the ratio of the amount of Chrysene compound to the amount of Benzo(a)anthracene compound for investigating the source of the PAHs concentrations (Notar et al., 2001; Tolosa et al., 2009). A magnitude less than 1 (Chr/BaA<1) indicates a source of fossil fuel combustion for PAHs and a magnitude bigger than 1 (Chr/BaA>1) indicates a fossil oil source for the PAHs (Notar et al., 2001). Based on the above criteria the source of PAHs concentrations in sediments at stations 1 and 2 were fossil fuel combustion and all other samples had petrogenic source (Table 2).

Sources of aliphatic hydrocarbons in the sediments from the study area were investigated using a set of indexes. Aliphatic hydrocarbons in the coastal sediments could be resulted from human activities or biological sources such as algae, bacteria, marine animals and terrestrial vascular plants (Tolosa et al., 2004; Meyers, 2003).

Figure 4 shows the distribution of n-alkanes in the sediment samples in the study area. The composition of n-alkanes in the sediments showed very similar pattern in most of the stations. High concentration and dominance of low carbon short-chain n-alkanes seen in the samples could indicate a fresh input of light oil (Tolosa. Et al., 2004).

The n-C16 index is computed from division of total n-alkanes concentrations to the concentration of n-C16. The amounts of n-C16 index less than 15 and more than 50 indicate that fossil or biological hydrocarbons are the sources of hydrocarbons, respectively (Clarke and Finely, 1973; Mirvakili et al., 2013). The n-C16 index of the sediments in the study area ranges from 9.8 to 23.5 and well below 50, indicating that they were contaminated with fossil hydrocarbons (Table 3).

The Carbon Priority index (CPI) is about 1 for crude oil hydrocarbons, while it varies from 3 to 6 for the hydrocarbons originated from vascular plants (Colombo et al., 1989). As shown in Table 3, the CPI index at all stations were approximately 1, indicating oil contamination in the sediment samples.

The Odd/Even index (Volkman et al., 1992) is the ratio of the total concentrations of odd carbon number alkanes to the total concentrations of even number ones. This ratio is about 1 for petrogenic hydrocarbons and varies between 8 and 10 for plant waxes. The Odd/Even index for the sediment in the study area (Table 3) were in the range of 0.6 to 1.3, indicating petrogenic hydrocarbons.

According to the results shown by all the presented indices, petrogenic inputs are the main source of hydrocarbons in the sediments in the study area. The results also indicated a fresh input of light oil.

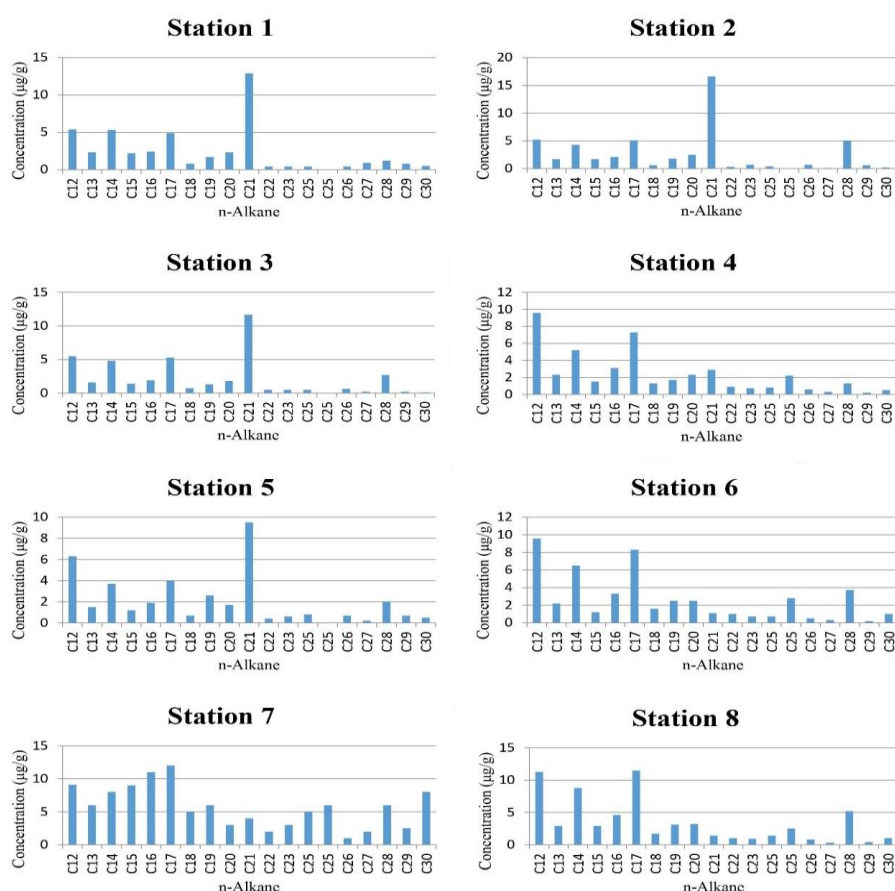


Figure 4. Values of n-alkanes concentrations in sediment samples ($\mu\text{g/g}$)

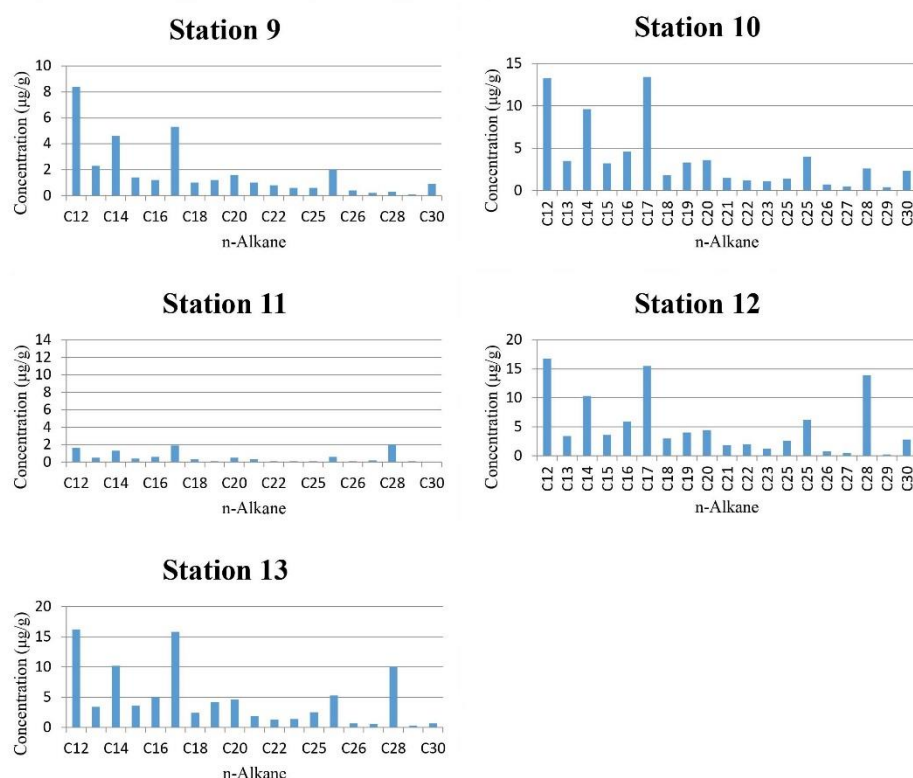


Figure 4. (continue). Values of n-alkanes concentrations in sediment samples ($\mu\text{g/g}$)

Table 3. Index quantities calculated for sediment samples

Station	CPI	n-C16	Odd/even
1	1.7	18.8	1.3
2	1.2	23.5	1.3
3	1.2	21.6	1.0
4	0.6	14.3	0.7
5	1.1	20.3	1.1
6	0.4	15.3	0.6
7	0.6	9.8	0.8
8	0.3	14.1	0.7
9	0.6	16.6	0.6
10	0.4	15.6	0.7
11	0.3	17.3	0.6
12	0.3	16.9	0.6
13	0.3	18.1	0.7

$$\text{CPI} = \frac{1}{2} \left(\frac{\sum C_{21-29} (\text{odd carbon})}{\sum C_{20-28} (\text{even carbon})} + \frac{\sum C_{21-29} (\text{odd carbon})}{\sum C_{22-30} (\text{even carbon})} \right)$$

CONCLUSIONS

The status and origin of aliphatic and aromatic hydrocarbons in Babolsar coastal area in the Caspian Sea were studied by measuring hydrocarbon concentrations in the seabed sediment sample. Maximum TPH concentration values were found in the sediments inside Babolrood River. In the coastal areas, TPH concentration was higher in sediments exposed to Babolrood River plume and the lowest TPH concentration was observed west of the study area where the plume effect was insignificant. The results showed that Babolrood River is the main source of petroleum hydrocarbon concentrations in the sediments of the study area. The oil pollution level in all river sediment samples and also in the ones from the coastal area under the effect

of the river plume, were moderate to high. In the sample from the western side of the river mouth, where the effect of the river plume was minimum, the oil pollution level was low. The results showed that the highest concentrations of 16 PAH compounds in the seabed sediment samples belonged to Naphthalene, Acenaphthylene, Fluorene, Phenanthrene, Fluoranthene and Pyrene. These compounds were observed at all stations. The results showed that pollution level with respect to \sum PAH concentration in all samples, except the one from control station in the west of the study area with no pollution, were moderate to significant. The results also showed petrogenic source for hydrocarbons in the sediments from the study area.

GRANT SUPPORT DETAILS

The present research did not receive any financial support.

CONFLICT OF INTEREST

The authors declare that there is not any conflict of interests regarding the publication of this manuscript.

LIFE SCIENCE REPORTING

No life science threat was practiced in this research

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