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Spatial distribution, enrichment and geo-accumulation of heavy metals in surface sediments near urban and industrial areas in the Persian Gulf

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ABSTRACT

Concentrations of Cd, Cr, Cu, Fe, Ni, Pb and Zn and geochemical controlling factors have been determined in surface sediment samples collected in the Persian Gulf (South of Iran) and were investigated using pollution indices including enrichment factor (EF), geo-accumulation index (Igeo) and contamination factor (Cf) distribution and its pollution status. Sediment samples were collected from 12 sites in a monitoring period of 4 months from September to December in 2014. The spatial distribution of heavy metals demonstrated that concentrations are much higher in the industrial areas in comparison with urban areas. Analytical results have been elaborated by using a geographical information system software to show metals accumulation areas. Based on the EF and Igeo, the order of heavy metal concentrations was Cd > Pb > Ni > Cr > Zn > Cu in industrial stations. For urban station, the order was as: Cd > Pb > Ni > Zn > Cu > Cr. EF and Igeo values showed that sediments were contaminated with Cd and Pb. Based on the EF, Cd was the heavy metal contaminant of most concern in urban and industrial surface sediments. Moreover, this study showed that Cd threshold concentrations to mostly be exceeded in the study area. Also, the concentration of heavy metals in the industrial areas was much higher than those of urban areas.

Keywords: Heavy metals; Sediment; Enrichment factor; Geo-accumulation index; Geographical information system; Persian Gulf

1. Introduction

Huge amounts of wastes find their ways into water and soil from numerous sources such as agricultural, industrial and domestic [1–6]. These wastes end up interacting with the soil system therefore, changing its physical and chemical properties. Sediment quality can be considered as suitable indicator so as to evaluate various pollutions in the environment [7–10]. Sediment acts as a medium for biogeochemical reactions influencing the food chain [9,11,12]. Organic and inorganic matters found in aquatic environment can move to sediment, therefore, this medium is a sink and provides history of different pollutions in the aquatic environment [13–15]. Soils contamination by numerous pollutants is nowadays one of the most important environmental issues. Following environmental pollution much attention has been devoted on trace metals over

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the recent decades due to heavy metals accumulated concentration in aquatic systems [16–18]. Heavy metals accumulate in sediments with high affinity to particle matters [19–21]. Soil is a dynamic natural resource for the human life and other organisms and due to its complex matrix is the principal receiver of the contaminants such as heavy metals. Therefore, determining the heavy metal concentrations in the sediments provides significant insights into the metal pollution in aquatic systems [22,23]. Because of development of industries and urbanization, especially in developing countries, soil contamination with heavy metals is now a great concern when they exceed certain prescribed limit. Heavy metals are released to soil and sediments through various routs including vehicle emission and industrial wastes [24–26]. The accumulation of heavy metals in soils is of increasing concern due to its potential health risks and detrimental effects on soil ecosystems [24,27–31]. Heavy metals in sediment have recently become a subject of many studies because of the serious risk represents for the environment and human health [32–34]. Metals are non-biodegradable and persistent environmental contaminants [35]. Thus, there should be concern about sediment pollution with heavy metals and potential health risks in coastal areas [36]. Parent rocks physical and chemical weathering, atmospheric deposition and wastewater discharge can affect the concentration of heavy metals in the sediments [18,37,38]. Through different mechanisms, adsorption, hydrolysis and co-precipitation of metal ions, a large quantity of heavy metals are deposited in the sediments. The accumulation and mobility of heavy metals in sediments controlled by various factors such as nature of the sediment particles, properties of adsorbed compounds, metal characteristics, redox reactions and biodegradation of sportive substance under specific conditions heavy metal in sediments are influenced by organisms and converted into organic complexes which can be hazardous to both human life and animals [34,39–41].

One of the techniques for comprehensive evaluation of the degree of—heavy metal accumulation in the soil is the use of pollution indices. There are many indicators such as contamination factor (Cf), the geo-accumulation index (Igeo) and enrichment factor (EF) to quantitatively determine the heavy metal contamination quantification [42–46]. In this study, we used geographical information system (GIS). GIS is a computer-based program that collects, analyzes and stores the information from the specific geographic location. It has been widely employed by many researchers to produce spatial distribution maps in relation to the accommodation of population, air pollution and climatic changes [47]. Considering the serious consequences brought by the heavy metal contamination in soil, it is urgent and necessary to control contamination. In this study, the concentrations of Cd, Cr, Cu, Pb, Ni and Zn in 12 surface sediment samples from urban and industrial area in Persian gulf were determined with the specific objectives (1) to investigate the spatial distributions of the heavy metals in the surface sediments; (2) to characterize the main pollutants, quantify the contamination levels and identify the potential sources by using the multivariate statistical analysis, EF methods, Igeo and Cf. Knowing contamination status of heavy metals in soils and identifying their environmental exposure hazards not only are important for soil pollution prevention, but also provide useful information for making decisions for remediation of the possible contaminated sites.

2. Materials and methods

2.1. Study area description

This study was conducted in sediments of Persian Gulf in the south and western parts of Bushehr province, Iran. Bushehr is in the south of Iran, with a long coastline onto the Persian Gulf. Its population was 1.163 million in 2016 and has 10 counties.

Bushehr climate is hot semi-arid climate (Köppen: BSh) with a precipitation pattern similar to Mediterranean climate. Asaluyeh city is the heartland of Iranian petrochemical industry in Bushehr and was considered for industrial area for sampling in this study.

2.2. Sampling

Six urban and six industrial sampling points (12 points) were selected along the coastal line of water during 4 months from September to December, 2014, in Persian Gulf. Totally 24 samples were taken and analyzed for heavy metal concentrations. The sampling locations were selected based on prevailing stressors on the environment including domestic and industrial places, as shown in Fig. 1. The depth of sediments sampling was 9.2 ± 2.2 in each sampling location. Sediment samples were collected through Ekman steel grab. The samples were immediately transferred to laboratory at –20°C and then dried in oven for 48 h [48,49]. After drying in an oven, sediment samples were ground by using a hand mortar followed by screening with a 0.5 mm sieve to remove large particles and debris to get fine fractions for chemical analyses.

2.3. Chemical analysis

Elemental analyses were performed in the Laboratory of Department of Environmental Health Engineering of Tehran University of Medical Science. 0.25 g subsamples were digested with a HNO_3–HF–$HClO_4$ mixture, and then the sample solutions were filtered, adjusted to a suitable volume with double deionized water. The total concentrations of Cd, Cr, Cu, Pb, Ni and Zn were analyzed using inductively coupled plasma-atomic emission spectrometry (ICP-AES, JY38S, Longjumeau, France). Finally, the concentration of the metals was reported in mg/kg dry weight in soil samples.

2.4. Statistical analysis

The results obtained were statistically analyzed using SPSS for Windows version 18 (SPSS Inc, Chicago, IL). Data analyses, including mean, maximum and minimum concentrations, and standard deviation, were performed. A GIS software (ArchView GIS 3.2a) was used in order to show metals accumulation areas in Persian Gulf [50,51].

3. Results and discussion

The spatial distribution of heavy metals concentration (Cd, Cr, Ni, Pb, Zn and Cu) throughout the case study is mapped in Fig. 1 and average heavy metals concentrations

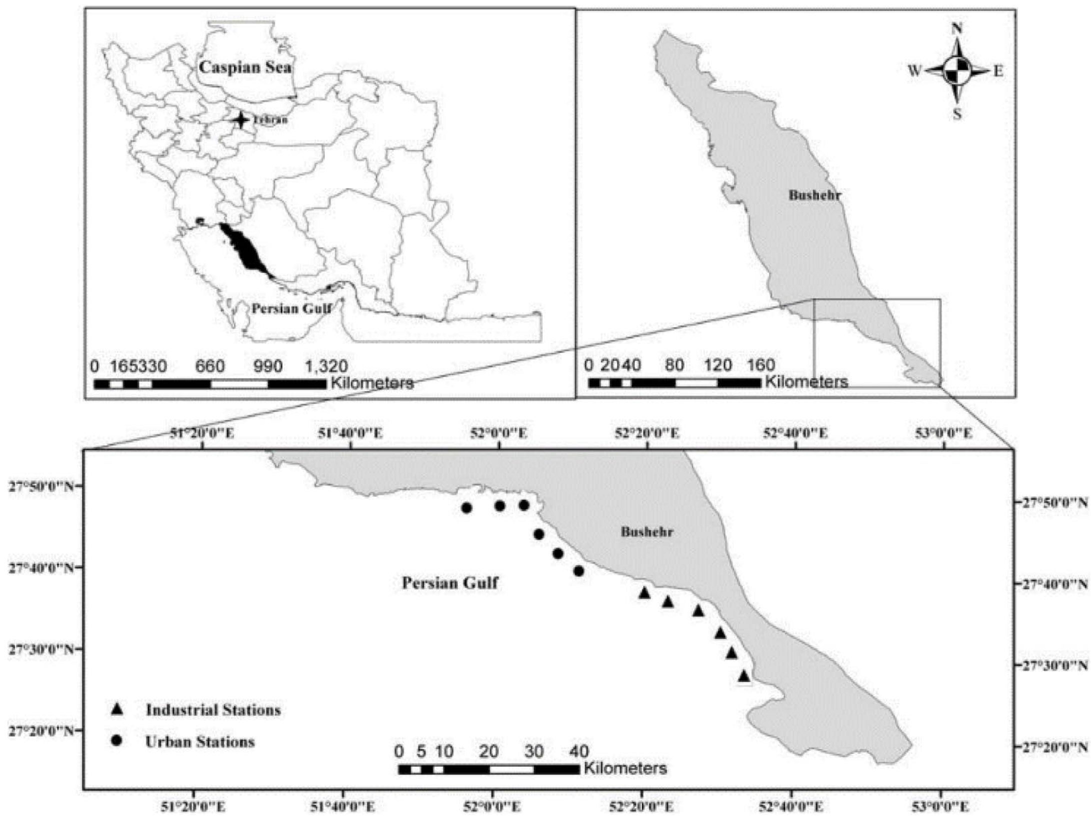

Fig. 1. Map of sampling stations.

Table 1
Average heavy metals concentrations (mg/kg dry weight) in the soils samples

Stations	Cd	Cr	Cu	Pb	Ni	Zn	Fe
1	1.75	54.32	31.74	22.36	52.32	87.5	53,274
2	1.94	57.41	33.56	27.89	50.74	87.32	45,047
3	1.34	72.56	33.43	56.7	51.33	95.34	58,126
4	2.14	79.63	35.2	24.33	49.45	94.31	57,000
5	2.04	60.23	41.7	34.37	57.76	95.2	47,600
6	1.87	58.97	42.56	40.39	45.03	98.76	49,748
Urban stations							
Minimum	1.34	54.32	31.74	22.36	45.03	87.32	49,600
Maximum	2.04	79.63	42.56	56.7	57.56	98.76	58,126
Average	1.85	63.85	36.36	34.34	51.1	93.07	51,799
7	2.5	93.24	43.67	60	67.31	101.2	47,350
8	3.2	87.97	37.6	77.5	75.9	105.4	55,493
9	3.38	105.61	51	79.9	77.3	101.7	59,000
10	3.01	97.38	33.97	70.31	60.43	102.9	62,000
11	2.96	89.34	35.5	56.99	59.71	95.5	57,000
12	2.64	93.21	37.84	50.33	62.3	93.89	49,000
Industrial stations							
Minimum	1.34	87.97	33.97	50.33	59.71	93.89	47,350
Maximum	3.38	105.61	51	79.9	77.3	105.4	62,000
Average	2.45	94.45	39.93	65.83	67.15	100	54,973
Mean of all stations	2.39	79.15	38.14	50.08	59.13	95.58	53,386

in the soils samples are listed in Table 1. As observed from Fig. 2, the concentration levels of heavy metals in urban area are lower as compared with industrial areas along the Persian Gulf. For the estimation and quantification of the degree of contamination in the soil samples, the following quantitative contamination indices were used to illustrate the concentration trends and also to allow rapid comparison among the determined values: contamination factor (Cf), the geo-accumulation index (Igeo) and EF.

3.1. Environmental indicators

The levels of pollution and its distribution were calculated through environmental indicators and compared to different valuable standards. The concentration of heavy metals in sediments was compared with local background area, where there are no industrial and domestic activities. To determine the levels of pollution in case study sediment, the average Turekian and Wedephol [53] indicator was employed.

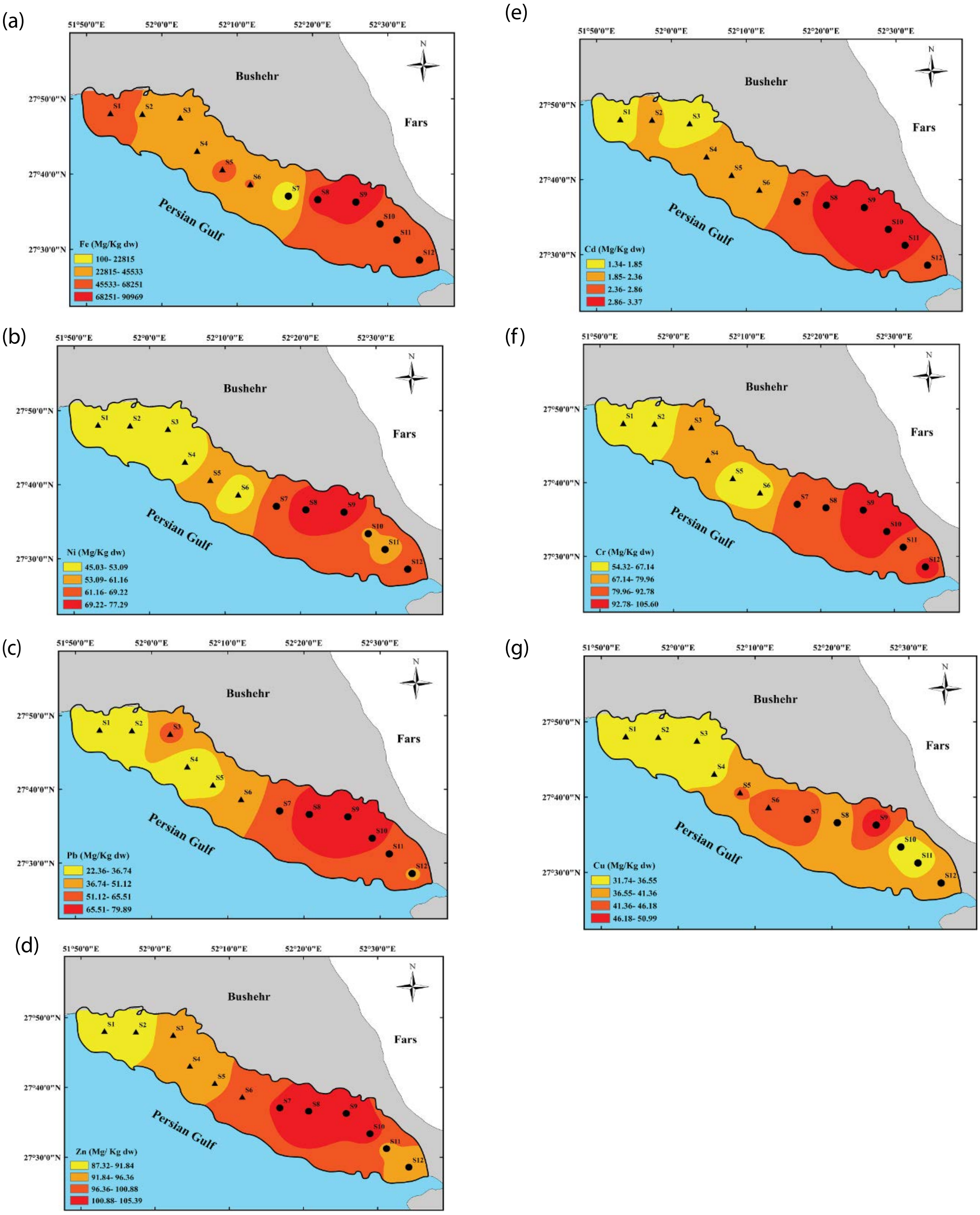

Fig. 2. Spatial distribution of Fe (a), Ni (b), Pb (c), Zn (d), Cd (e), Cr (f), and Cu (g) in surface sediments of the Persian Gulf.

The concentration levels of different heavy metals according to average shale are listed in Table 2.

3.2. Enrichment factor

In order to provide information for better understanding of sediment quality in the sediments regarding heavy metals, levels of metal pollution were evaluated using different techniques. One of them is "enrichment factor (EF)" which is a powerful tool to distinguish between anthropogenic and naturally occurring sources of heavy metals. EF technique is used in the area of atmospheric aerosols, soil, sediments and solid wastes to determine the degree of modification in the composition [52]. For this study, we chose Fe as the element, because the natural concentration of Fe in the core sediments were more uniform than of Al. EF is expressed by the following equation:

$$EF = \frac{(M/Fe)_{sample}}{(M/Fe)_{background}} \tag{1}$$

where $(M/Fe)_{sample}$ is the metal/Fe ratio in the sample of interest and $(M/Fe)_{background}$ is the natural background value of the metal/Fe ratio [52].

Birch et al. [11] divided contamination into different categories based on EF values. EF < 1 demonstrates "no enrichment", EF < 3 is "minor enrichment", EF = 3–5 is "moderate enrichment", EF = 5–10 is "moderately severe enrichment", EF = 10–25 is "severe enrichment", EF = 25–50 is "very severe enrichment" and EF > 50 is "extremely severe enrichment". In this study, EF values were 14.01 for Cd industrial station (severe enrichment) and 9.44 for urban station (moderately severe enrichment), EF values are 0.9 for Cr industrial station (no enrichment) and 0.64 for urban station (no enrichment), EF values are 0.76 for Cu industrial station (no enrichment) and 0.74 for urban station (no enrichment), EF values are 2.81 for Pb industrial station (minor enrichment) and 1.55 for urban station (minor enrichment), EF values are 1.15 for Ni industrial station (minor enrichment) and 0.93 for urban station (no enrichment), EF values are 0.9 for Zn industrial station (no enrichment) and 0.89 for urban station (no enrichment). The different categories of EF are described in Table 3 [52].

3.3. Contamination factor

For describing the contamination of a given toxic substance in a basin, a contamination factor was used by Hakanson [40]. Contamination factor (Cf) is a ratio of the concentration of the element in samples to preindustrial reference value for the element.

$$Cf = \frac{C}{C_p} \tag{2}$$

$$Cd = \sum Cf \tag{3}$$

According to Hakanson [40], Cf has been classified into four groups: Cf < 1 low contamination factor, 1 ≤ Cf < 3 moderate contamination factor, 3 ≤ Cf < 6 considerable contamination factor and Cf > 6 very high contamination factor as shown in Table 4. In the calculation of Cf, the average shale values and/or average crustal abundance were used [53,54].

The Cf values of Cd, Cr, Cu, Pb, Ni, Zn were calculated based on using the measured concentration of each metal. Accordingly, metal levels existed in order of Cd > Pb > Ni > Zn > Cr > Cu in the industrial station and Cd > Pb > Ni > Zn > Cu > Cr in urban stations in the Persian

Table 2
Concentration of different heavy metals according to average shale

	Average shale
Cd	0.18
Cr	90
Cu	45
Pb	20
Ni	50
Zn	95
Fe	47,000

Table 3
Seven classes of enrichment factor

Value enrichment	EF
No enrichment	<1
Minor enrichment	<3
Moderate enrichment	3–5
Moderately severe enrichment	5–10
Severe enrichment	10–25
Very severe enrichment	25–50
Extremely severe enrichment	≥50

Table 4
Classification of Hakanson for Contamination factors (Cf) and degree of contamination (Cd) of surface sediments

Contamination factor of sedimentation	Cf value	Quality of sedimentation	Cd Value
Low contamination factor	<1	Low contamination degree	<6
Moderate contamination factor	1 ≤ Cf < 3	Moderate contamination degree	6 ≤ Cd < 12
Considerable contamination factor	3 ≤ Cf < 6	Considerable contamination degree	12 ≤ Cd < 24
Very high contamination factor	Cf > 6	Very high contamination degree	Cd > 24

Gulf (Table 5). In industrial stations, the Cf values showed very high contamination for Cd, moderate contamination for Pb, Ni, Zn and Cr and low contamination for Cu. In urban stations, the Cf values of Cd indicated very high contamination, Pb and Ni indicated moderate contamination and Zn, Cu and Cr showed low contamination. The maximum Cf value was found for Cd in an industrial station.

Comparison between the Cd and Cf shows that the contamination of the surface sediments in the industrial station of the Persian Gulf is dominated for Cd, Pb, Ni, Zn and to a lesser extent for Cr, Cu. Also, compared with the literature values reported for the heavy metal in sediments, the values of heavy metal concentrations were lower than that reported by previous studies. A comparison of metal values in sediments reported in other studies is shown in Table 7. Comparison of enrichment factor, contamination factor and geo-accumulation index of metals in the Persian Gulf is given in Table 8.

3.4. Geo-accumulation index

To estimate the enrichment of metal concentrations above background or baseline concentrations, a common approach is to calculate the geo-accumulation index (Igeo) as proposed by Muller [55]. Sediment Igeo is the quantitative measure of heavy metal pollution in aquatic sediments. This index is basically a single-metal approach to quantify metal pollution in sediments when the concentration of toxic heavy metal is 1.5 or several times greater than their lithogenic background values [56].

$$\text{Igeo} = \log_2\left(\frac{C_n}{1.5B_n}\right) \quad (4)$$

where C_n is the measured concentration of the element n and B_n is the geochemical background value element n in average crust. The factor 1.5 is used to compensate possible variations which may be attributed to lithologic variations in the sediment. Geochemical background values of Fe and Mn were taken from Turekian and Wedepohl [53]. Muller [55] and Zhang et al. [57] introduced seven categories for this indicator, which the highest value is at least 10 magnitudes of orders higher than the background value (Table 6).

In the present research, Igeo indicator for different heavy metals in urban areas was found to be: Cd (2.75, moderately polluted to highly polluted), Cr (–1.09, unpolluted), Cu (–0.9, unpolluted), Pb (0.11, unpolluted to moderately polluted), Ni (–3.34, unpolluted) and Zn (–0.6, unpolluted). As for industrial areas, the aforementioned indicator for seven heavy metals was as follows: Cd (3.44, highly polluted), Cr (–0.5, unpolluted), Cu (–0.7, unpolluted), Pb (1.11, moderately polluted), Ni (–0.16, unpolluted) and Zn (–0.5, unpolluted) (Figs. 3 and 4).

Table 6
Degree of pollution in sediments based on geochemical accumulation

Soil quality	Igeo class	Number of measurement
Unpolluted	0	≤0
Unpolluted to moderate polluted	1	0–1
Moderate polluted	2	1–2
Moderate polluted to high polluted	3	2–3
High polluted	4	3–4
High polluted to extremely polluted	5	4–5
Extremely polluted	6	>5

Table 5
Contamination factors (Cf) and degree of contamination (Cd) of surface sediments in the Persian Gulf

Station	Cf						Cd
Urban	Cd	Cr	Cu	Pb	Ni	Zn	
1	9.72	0.6	0.7	1.11	1.04	0.92	14.11
2	10.77	0.63	0.74	1.39	1.01	0.91	15.48
3	7.44	0.8	0.74	2.83	1.02	1	13.85
4	11.88	0.88	0.78	1.21	0.98	0.99	16.75
5	11.33	0.66	0.92	1.71	1.15	1	16.8
6	10.38	0.65	0.94	2.01	0.9	1.03	15.94
Mean of urban station	10.26	0.7	0.8	1.71	1.02	0.97	15.49
Industrial							
7	13.88	1.036	0.97	3	1.34	1.06	21.3
8	17.77	0.97	0.83	3.87	1.51	1.1	26.09
9	18.77	1.17	1.13	3.99	1.54	1.07	27.69
10	16.72	1.082	0.75	3.51	1.2	1.08	24.36
11	16.44	0.99	0.78	2.84	1.19	1	23.27
12	14.66	1.03	0.84	2.51	1.24	0.98	21.29
Mean of industrial station	16.37	1.04	0.88	3.29	1.34	1.05	24

Table 7
Comparison among geochemical background values (B_n) for metal in sediments reported in other studies

Element	This study	Taranto gulf (Alessandro Buccolieri)	Inner ezmir bay (Ebru Yeşim Özkan)	Milos bay	B_n average shale
Fe	4,700	26,313	47,000	34,000	47,200
Cr	95	81	95	119	95
Cu	45	47	45	51	45
Zn	95	97	95	325	95
Ni	50	57	68	61	68
Cd	0.18	–	0.17	–	0.18
Pb	20	59	20	151	20

Table 8
Comparison of enrichment factor, contamination factor and geo-accumulation index of metals in the Persian Gulf

Heavy Metal	EF	Cf	Igeo
		Urban stations	
Cd	Moderately severe enrichment	Very high contamination	Moderately polluted to highly polluted
Cr	No enrichment	Low contamination	Unpolluted
Cu	No enrichment	Low contamination	Unpolluted
Pb	Minor enrichment	Moderate contamination	Unpolluted to moderate polluted
Ni	No enrichment	Moderate contamination	Unpolluted
Zn	No enrichment	Low Contamination	Unpolluted
		Industrial stations	
Cd	Severe enrichment	Very high contamination	High polluted
Cr	No enrichment	Moderate contamination	Unpolluted
Cu	No enrichment	Low contamination	Unpolluted
Pb	Minor enrichment	Considerable contamination	Moderate polluted
Ni	Minor enrichment	Moderate contamination	Unpolluted
Zn	No enrichment	Moderate contamination	Unpolluted

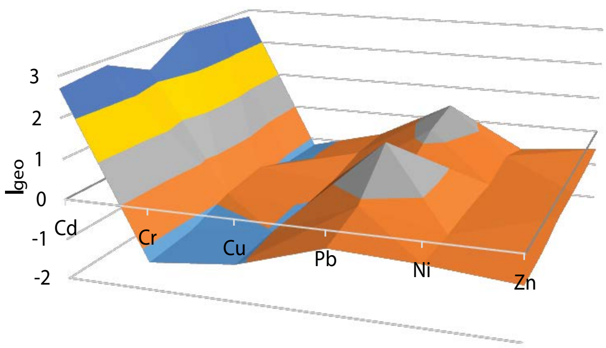

Fig. 3. Spatial distribution of Igeo in the surface sedimentation of the Persian Gulf (near urban areas).

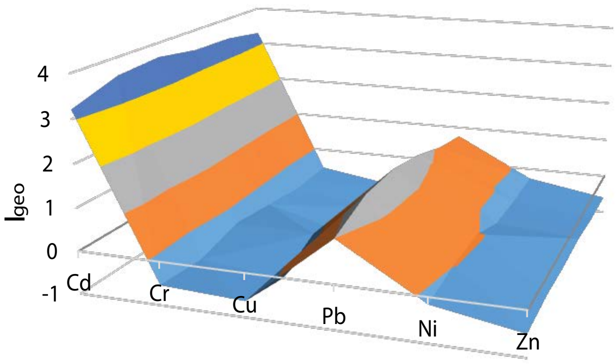

Fig. 4. Spatial distribution of Igeo in the surface sedimentation of the Persian Gulf (near industrial areas).

The results of the present research are compared with the results of other studies in literature. Ganugapenta et al. [58] assessed the heavy metal pollution from the sediment samples of Tupilipalem Coast, southeast coast of India. The Igeo showed that the surface sediment of the Tupilipalem Coast was highly contaminated with Fe, Mn, Cr, Cu, Ni, Pb and Zn. The computation of EF indicated a significant enrichment for Pb, Zn and Cd and a moderate enrichment for Cr, Cu and Ni [58]. Bazzi [59] determined the level of heavy metal pollution in surface sediments of the Gulf of Chabahar, Iran. In winter, the concentration of heavy metals including Cu, Pb, Zn, Cd and Ni was more than of the summer season. Generally, concentration of heavy metals was lower than the quality standards for sediments and water. However, between different studied stations, higher concentrations of heavy metals were found in stations nearby the fishing ports [59]. Pazi [60] studied heavy metal (Fe, Mn, Ni, Cu, Zn, Pb, Hg, Cr, Al and As) contamination in Candarli Gulf sediment, Eastern Aegean Sea. Concentrations of heavy metals in surface sediment varied from 1.62% to 3.60% for Fe, 0.38%–2.53% for Al, 173–1,423 for Mn, 8–100 for Ni, 3–46 for Cu, 55–119 for Zn, 16–138 for Pb, 0.2–6.3 for Hg, 16–71 for Cr and 11–37 mg/kg for As [60]. Nasr et al. [61] studied heavy metal pollution in bottom sediments of Aden Port, Yemen and reported the range and average concentrations measured were 138.23–658.87 (335.5) for Mn, 21.85–263.49 (128.59) for Zn, 8.06–111.00 (19.89) for Cu, 14.8–138.06 (77.28) for Pb, 13.8–33.64 (23.97) for Co, 17.00–233.93 (82.19) for Cr and 16.17–48.07 (34.54) for Ni in mg/kg. Generally, the Cf of heavy metals in the present study could be arranged as follows: Pb > Cr > Zn > Co > Ni > Cu > Mn. Krika and Krika [62] evaluated the heavy metals pollution in water and sediments of Djendjen River, North Eastern Algeria. The distribution of heavy metals in water and sediment was as: Ni > Zn > Cd > Cu and Zn > Ni > Cu > Cd, respectively. The Igeo values showed that Cd was highly accumulated compared with the other metals. Cf confirms that the sediment samples have been moderate in terms of all investigated metals contamination. The values have been above one (>1), signifying an advanced reduction of the sediment quality [62]. In another study the heavy metal pollution (Zn, Cr, Cu, As, Cd, and Pb) in the sediment of the main tributaries of Dongting Lake, China were assessed. According to the Igeo and EF, heavy metals pollution was very high in the sediment of lake, and Cd was the dominant pollutant, indicating considerable potential ecological risks [63]. Assessment of heavy metals (Cr, Co, Ni, Cu, Zn, As, Cd, Pb and Hg) contamination and ecological risks in 200 sediment samples along the coast of Korea was performed. Average concentrations were 58.3 mg/kg for Cr, 10.2 mg/kg for Co, 24.3 mg/kg for Ni, 36.5 mg/kg for Cu, 122 mg/kg for Zn, 9.1 mg/kg for As, 0.25 mg/kg for Cd, 35 mg/kg for Pb and 0.046 mg/kg for Hg, respectively, in sediment samples. The metal indices revealed that the Korean coast sediments were moderately contaminated with Cu, Zn, As, Cd, Pb and Hg metals. The average of metal enrichment values declined in the order of Cu > Hg > Cd > Zn > As > Pb > Co > Cr > Ni [64].

4. Conclusions

The main objective of present research was to quantitatively ascertain the pollution levels of heavy metals in urban and industrial areas based on indicators EF, Cf and Igeo. The results indicated that concentration levels of Cd, Cr, Cu, Pb, Ni and Zn in industrial areas are as high as domestic areas. The reason for the facts can be attributed to establishment of gas refinery, petrochemical companies near the stations 8, 9 and 10, generating much heavy metals pollution. Anyway, pollution above the cadmium metal in most urban and industrial stations may be due to the entry of urban and industrial wastewater, rivers connected to the sea, leakage from tanker ships, fishing vessels and hunting vessels, shipbuilding industry, naval ports and ships. The results indicated that the concentration levels of heavy metals are reduced as number and density of companies placed in the case study are lowered (stations: 11 and 12). It is hypothesized that the high concentration levels of Cu in sampling stations 5 and 6 were attributed to discharging Portland factory wastewater to environment near the stations. Therefore, it can be concluded that gas and oil refinery and petrochemical activities play important roles in heavy metal pollutions in Asaluyeh city and the Persian Gulf sediments.

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Conflict of interest

The authors of this article declare that they have no conflict of interests.

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