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Contamination and Risk Assessment of Potentially Toxic Elements in Coastal Sediments of the Area between Al-Jubail and Al-Khafji, Arabian Gulf, Saudi Arabia

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Abstract: Coastal environments need continuous environmental risk assessment, especially with increasing coastal development and human activities. The present work evaluates the distribution, contamination, and environmental risk of potentially toxic elements (PTEs) in coastal sediments between Al-Jubail and Al-Khafji cities along the Arabian Gulf, Saudi Arabia, and documents the influence of background references applied in pollution indices. Thirty-two sediment samples were collected for analysis of Ni, Cu, Cr, As, Zn, Pb and Hg using ICP-AES. The ranges of PTEs (mg/kg) were in the following order: Cr (3.00–20.0), Ni (2.00–32.0), Zn (2.00–14.0), As (2.00–4.00), Pb (1.50–5.00), Cu (1.00–5.00), and Hg (0.50–1.00). The coastal sediments show severe enrichment with As and Hg, and no to minor enrichment and a low contamination with Cr, Cu, Cr, Zn, and Pb. Based on sediment quality guidelines, concentrations of Cu, Pb, Zn, As, and Cr do not represent a concern for benthic communities, while Ni and Hg show a risk for benthic communities in four and 17 sampled areas, respectively. Multivariate analysis indicated a geogenic source for Zn, Cr, Cu, Ni, and Pb, mixed natural and anthropogenic sources for As, and an anthropogenic source for Hg, mostly from oil pollution, sewage, and industrial effluents spreading near Al-Jubail industrial city.

Keywords: risk assessment; background references; potentially toxic elements; ICP-AES; Arabian Gulf; Saudi Arabia

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1. Introduction

The worldwide coastal areas have been subjected to a strong pressure due to human settlement and the high amount of industrial and residential wastes, causing problems for marine organisms and human health [1–3]. PTEs enter the coastal environment through natural and anthropogenic sources, especially with the rapid development over the past decades [4,5]. However, PTEs can be remobilized to the water column and accumulate in marine organisms in thousands of times through transfer in the marine food chain leading to potential ecological risks and a huge threat to public safety and health [6,7].

The lack of environmental quality standards and the lack of information related to the PTE levels presented in these environments before human settlement and industrial development were the main obstacles and difficulties related to assess the anthropogenic impacts on coastal ecosystems [8]. To evaluate the PTE enrichment or the degree of contamination in sediment compared to reference reflects a natural condition; pollution indices, such as the enrichment factor, contamination factor, and pollution load index were frequently used [9,10]. The background level refers to the normal concentrations of metals found in environments without any anthropogenic intervention [11]. The use of contamination indices to evaluate the impact of pollutants in marine sediments requires background references, which are not always available at the local scale. Thus, most of the worldwide studies use backgrounds defined as global references.

The coastal zones of the Arabian Gulf have been subjected to intensive monitoring studies [12–23]. The shoreline between Al-Jubail and Al-Khafji cities is primarily made up of sand, with some artificially and naturally occurring rocky areas, which are mostly bioeroded by different bivalves, annelids, sponges, gastropods, and barnacles [24–26]. The shoreline sediments still lack the information about contamination and risk assessment of PTEs, and the impacts of the natural and anthropogenic substances on the marine benthic communities and human health. Therefore, the present work aims to document the contamination and spatial distribution of Ni, Cu, Cr, As, Zn, Pb and Hg in marine sediments near one of the most industrialized areas along the Arabian Gulf, and to evaluate the ecological risk of PTEs on benthic assemblages, using different background references in contamination indices.

2. Materials and Methods

2.1. Study Area and Sampling

The study area is located between Al-Jubail and Al-Khafji cities along the Arabian Gulf coast, between N27°00′84″–N28°18′26″ and E49°40′00″–E48°31′37″ (Figure 1). It is assumed to be nearby the Al-Jubail industrial city, which is highly populated with intense petrochemical industrial activities, desalinization plants, and fertilizer and cement factories [16,19,27]. The coastal sediment in the study area contains both biogenic and terrestrial components. The biogenic quotient consists of fragments and whole foraminiferal tests, bivalves, gastropods, echinoids, ostracods, corals, and algae, which drifted from offshore to the beach during storms and tides (landward migration). The terrestrial part, composed of quartz grains and rock fragments, came to the beach from the hinterland Quaternary sediments. Sediment samples were collected from thirty-two sampling sites from the intertidal zone at 0.15 to 0.50 m water depth (3 replicates from each site). Sediments were collected in plastic bags for storage and transportation. The sediments were dried to constant mass in a 115 °C drying oven and sieved using a nest of sieves (>500 µm, 500–250 µm, 250–125 µm, 125–63 µm, and <63 µm).

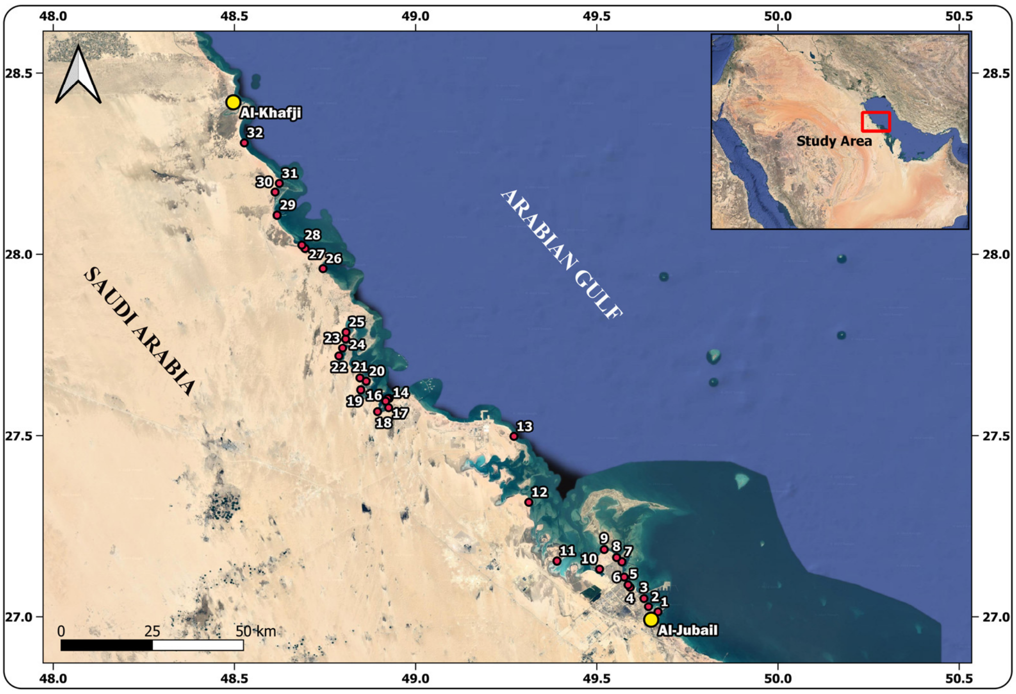

Figure 1. Location of the study area and sample sites along the Arabian Gulf coast.

2.2. Analytical and Assessment Methods

The dried sediments were ground using a mortar and pestle to a particle size less than 150 µm. The dried, ground sediments from each site were digested. Approximately 0.50 g of each prepared sample is digested with aqua regia (a mixture composed of one mole of nitric acid and three moles of hydrochloric acid) for 45 min, in a graphite heating block. After cooling, the resulting solution is diluted to 12.5 mL with deionized water. Analysis of Ni, Cu, Cr, As, Zn, Pb and Hg were performed in the ALS Geochemistry Lab, Jeddah branch, Saudi Arabia using a Perkin-Elmer Optima 3100XL axial viewing inductively coupled plasma-atomic emission spectroscopy (ICP-AES, Waltham, MA, USA) equipped with a cyclonic spray chamber and a GemTip cross-flow nebulizer. The detection limits in ICP-AES for the analysis of PTEs were between 1.0 and 10,000 mg/kg. Table S1 presents the location (GPS) of the sampling sites and the PTE concentrations (mg/kg). A complete description of this instrument, as well as instrumental parameters and conditions for the determined PTEs in this study, are detailed elsewhere [28,29].

Validation of the ICP–AES technique was performed with regard to the linearity, limit of detection (LOD), and limit of quantification (LOQ). Three samples were analyzed in duplicate to verify the precision of analysis. The LOD value was the concentration that corresponds to three times the standard deviation of the measurements for the blank solutions divided by the slope of calibration curves for each element, while the LOQ value was the concentration that corresponds to ten times the standard deviation of the measurements for the blank solutions divided by the slope of calibration curves for each element [30,31]. The relative standard deviations (RSD%) for all elements were lower than 13.5%, demonstrating that the method offered good precision [28]. The relative recovery values (R%) ranged between 80–120%, demonstrating that the method offered good accuracy. The calibration procedure was performed by the preparation of a stock standard solution of all investigated elements with concentrations of 1000 mg/kg. The single element solutions of each of the investigated elements with concentrations 1, 5, and 10 mg/kg, respectively, was prepared from stock solutions by dilution with tridistilled water. The ALS Geochemistry Laboratory has established a sound quality control/quality assurance experience and protocol over many similar studies throughout the years.

The enrichment factor (EF), contamination factor (CF), and pollution load index (PLI) were used to evaluate the sediment contamination with particular PTEs [32]. The sediment quality guideline (SQG) procedure was used to predict the adverse effects produced by polluted sediments on aquatic organisms [33,34]. These indices are classified in Table 1, and calculated according to the following formulas [9,11,35,36]:

$$\mathrm{EF} = (\mathrm{M/X})_{\mathrm{sample}} \div (\mathrm{M/X})_{\mathrm{background}}$$

$$\mathrm{CF} = \mathrm{C_o} \div \mathrm{C_b}$$

$$\mathrm{PLI} = (\mathrm{CF_1} \times \mathrm{CF_2} \times \mathrm{CF_3} \times \mathrm{CF_4} \ldots \times \mathrm{CF_n})^{1/n}$$

$$\mathrm{m-ERM-Q} = \frac{\sum_{i-1}^{Q}\left(\frac{C_i}{\mathrm{ERM}_i}\right)}{n}$$

where M, C_o, and C_i are the analyzed metal; X is the concentration of a normalizer element (Fe); C_b is the referred metal on the background; and PLI is calculated using the n-th root of the product of n contamination factors (CFs) for the tested PTEs. ERM_i is the ERM value of metal i, and n is the number of metals. The chemical concentrations in the 19th percentile are known as effects range—low (ERL), and those in the 50th percentile are effects range—median (ERM) [34]. The ecological risk assessment (ERA) of Long et al. [33] was used to evaluate the hazard of the contaminated sediments, considering the mean ERM quotient (m-ERM-Q). Thus, the m-ERM-Q represents the effects of multiple anthropogenic contaminations. Two of the famous and widely used background references were applied herein to document their difference on pollution indices and, consequently, the risk assessment [37,38]. Hierarchical clustering analysis (HCA), Pearson's correlation coefficients, and principal component analysis (PCA) were applied as multivariate statistical tools (using Microsoft Excel 2016 and SPSS 16.0 statistical software) to identify the possible sources of PTEs in the investigated sediment.

Table 1. Classification of the indices applied in this work.

EF	EF < 1	no enrichment
	EF < 3	minor enrichment
	EF = 3–5	moderate enrichment
	EF = 5–10	moderately severe enrichment
	EF = 10–25	severe enrichment
	EF = 25–50	very severe enrichment
	EF > 50	extremely severe enrichment
CF	CF < 1	low contamination
	$1 \leq$ CF <3	moderate contamination
	$3 \leq$ CF < 6	considerable contamination
	CF ≥ 6	very high contamination
PLI	PLI < 1	unpolluted
	PLI > 1	polluted
m-ERMQ	m-ERMQ < 0.1	9% probability of toxicity
	$0.1 \leq$ m-ERMQ < 0.5	21% probability of toxicity
	$0.5 \leq$ m-ERMQ < 1.5	49% probability of toxicity
	$1.5 \leq$ m-ERM-Q	76% probability of toxicity

3. Results and Discussion

3.1. Distribution of PTEs

The average concentrations of PTEs (mg/kg) had the following descending order: Ni (11.76) > Cr (8.68) > Zn (6.18) > Pb (2.57) > Cu (2.44) > As (2.38) > Hg (0.76). Table 2 illustrated the minimum, maximum, and average (mg/kg, dry weight) of the investigated PTEs along with those from background references and other coastal sediments. The average value of Hg exceeds those from Earth's crust and Gulf of Suez coastal sediment [37–39]. Differently, As, Cu, Ni, Pb, and Zn reported average values less than Earth's crust backgrounds and Mediterranean Sea coastal sediment [40]. The average Cr value exceeds those recorded from coastal sediment of the Mediterranean Sea (Egypt) and Ras Abu Ali Island, Saudi Arabia [40,41]. Figure 2 presents the distribution of the PTEs in the sampling sites. Q-mode HCAour clusters the thirty-two samples into three clusters (Figure 3). The first cluster includes 17 samples (S3, S7, S8, S10, S13–S16, S18, S22–S24, S26–S28, S32), the second cluster accounts for 11 samples (S1, S2, S4–S6, S9, S11, S12, S19, S25, S29), and the third cluster includes four samples (S17, S20, S30, and S31). The first cluster reported the lowest values of all PTEs, indicating natural factors and implying the role of clayey sediment in the accumulation and adsorption of the PTEs [4,22,42]. Samples of the second cluster account for the highest values of Pb, Hg, and As, while samples of the third cluster reported the highest concentrations of Cu, Cr, Ni, and Zn.

Table 2. Minimum, maximum and average values of PTEs (mg/kg, dry weight) in the study area, along with those from background references and other coastal sediment.

Location and References		As	Cr	Cu	Ni	Pb	Zn	Hg
Present study	Minimum	2.00	3.00	1.00	2.00	1.50	2.00	0.50
	Maximum	4.00	20.0	5.00	32.0	5.00	14.0	1.00
	Average	2.38	8.68	2.44	11.76	2.57	6.18	0.76
Earth's crust [37]		13.0	90.0	45.0	68.0	20.0	95.0	0.40
Continental crust [38]		1.80	100	55	75.0	12.50	70.0	0.08
Suez Bay, Gulf of Suez, Egypt [39]			8.98	1.66	5.58	2.78	3.96	0.27
Rosetta, Mediterranean Sea, Egypt [40]		298	0.18	24.57	481	385	183	
Ras Abu Ali Island, Saudi Arabia [41]		2.47	7.86	4.14	13.0	3.50	6.89	

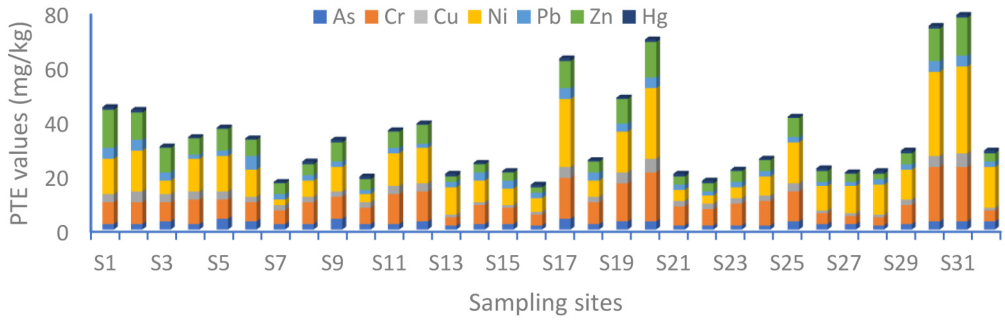

Figure 2. Distribution of the PTEs in the coastal sediment between Al-Jubail and Al-Khafji.

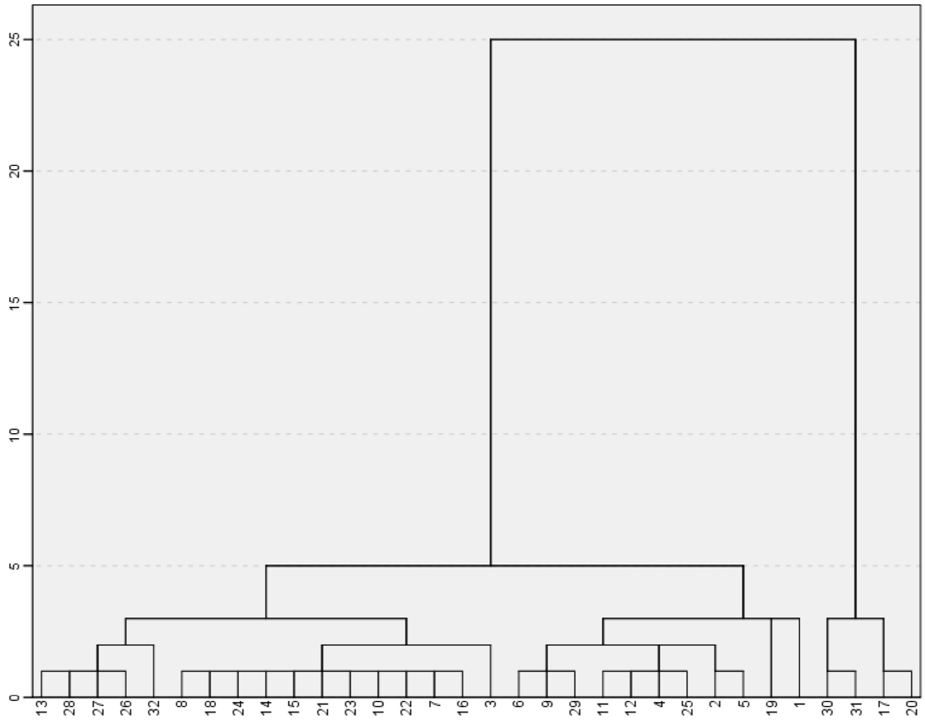

Figure 3. The Q-mode hierarchical clustering analysis of the investigated 32 sediment samples.

3.2. Contamination and Risk Assessment of PTEs

In order to estimate the anthropogenic effects of marine sediments through enrichment factors, the choice of a background metal concentration is crucial. The main drawback of using crustal average values is that it ignores natural geochemical variability, which can result in the detection of erroneous anomalies or the failure to detect anomalous concentrations above the pristine local background at all [43]. Average values of Cr, Cu, Cr, Zn, and Pb were less than 3.00 (Table 3), indicating no to minor enrichment, and may be derived entirely from natural crustal materials with minor anthropogenic effects for Pb [11,44–46]. These PTEs show minor difference in EF average values among the two used background references (Figure 4).

However, arsenic (As) revealed severe enrichment in EF average values (14.50 and 16.58), and show minor differences based on the background values (Table 3, Figure 4). Moreover, the average values of EF for Hg varied from 16.73 to 21.40, indicating severe enrichment, mostly derived from anthropogenic effects. The considerable difference between the two averages is attributed mostly to the difference in the abundance of mercury in the Earth's crust (0.40 mg/kg for Turekian and Wedepohl [37]; and 0.08 mg/kg for Taylor [38]), as well as the difference in Fe content as a normalizing element between the two backgrounds.

Table 3. The average values of enrichment factor (EF) and contamination factor (CF), based on background values.

Indices and Background References		Pb	Zn	Ni	Cu	Cr	As	Hg
EF	Turekian and Wedepohl [37]	1.27	0.61	2.01	0.47	0.90	14.50	21.40
	Taylor [38]	2.27	0.99	1.89	0.48	0.90	16.58	16.73
CF	Turekian and Wedepohl [37]	0.13	0.07	0.20	0.05	0.10	1.40	2.47
	Taylor [38]	0.21	0.09	0.16	0.04	0.09	1.32	1.38

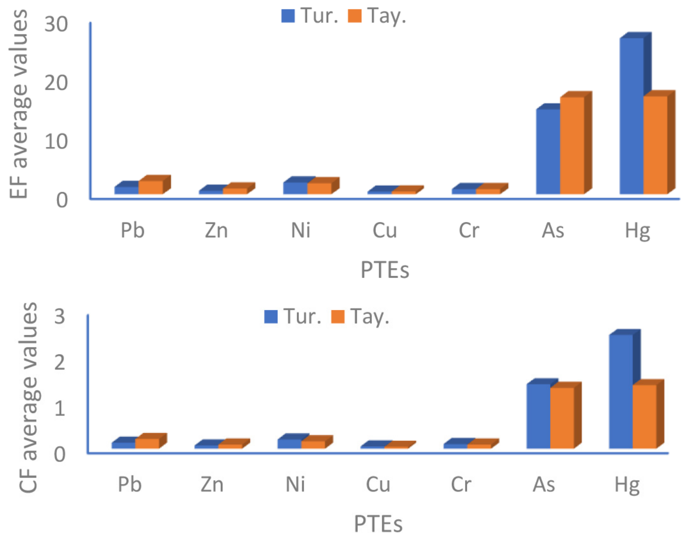

Figure 4. Distribution of EF and CF average values of PTEs based on background values [37,38].

The coastal sediment in the area between Al-Jubail and Al-Khafji shows a low contamination factor for Pb, Zn, Ni, Cu, and Cr (average CF < 1.00), with minor differences among the used backgrounds (Table 3, Figure 4). Moreover, As and Hg show average CF values varied from 1.32 to 1.40 and from 1.38 to 2.47, respectively, based on the applied backgrounds, indicating a moderate contamination factor [35]. The pollution load index (PLI) is used to assess HM contamination at a particular site [47,48]. Results of PLI indicate unpolluted sediments (PLI < 1.00), mostly due to low contamination factors for all PTEs (Table 3). Average values of PLI varied from 0.17 to 0.19, based on used background values [37,38]. The higher PLI values were recorded in S17, S20, S30 and S31 (Figure 5). However, the results of pollution indices (EF, CF, and PLI) support the results of the Q-mode HCA and indicate that samples of the first cluster were not polluted, while samples of the second and third clusters show some enrichment of anthropogenic effects, especially for Hg and As.

Table 4 presents the range of ERL and ERM values of the sediment quality guideline in the seven PTEs [33], and the percentages of samples within the ranges of the SQG. Cu, Pb, Zn, As, and Cr present the 100% of measurements below the ERL, indicating that the studied coastal sediments do not pose a risk to the benthic communities, or the adverse effects are rarely observed as a result of the presence of these metals. However, Ni presented 87.5% below the ERL, and 12.5% between the ERL and ERM, which suggests that the presence of Ni could represent a risk for benthic communities in four sampled areas (S17, S20, S30, and S31). On the other hand, 47% of the Hg values located between ERL and ERM, and 53% over the ERM imply that Hg represents an evident risk for benthic communities in 17 sampled areas [8]. Based on SQG results, the m-ERMQ were 0.1, suggesting a 9% probability of toxicity for benthic organisms in the study area.

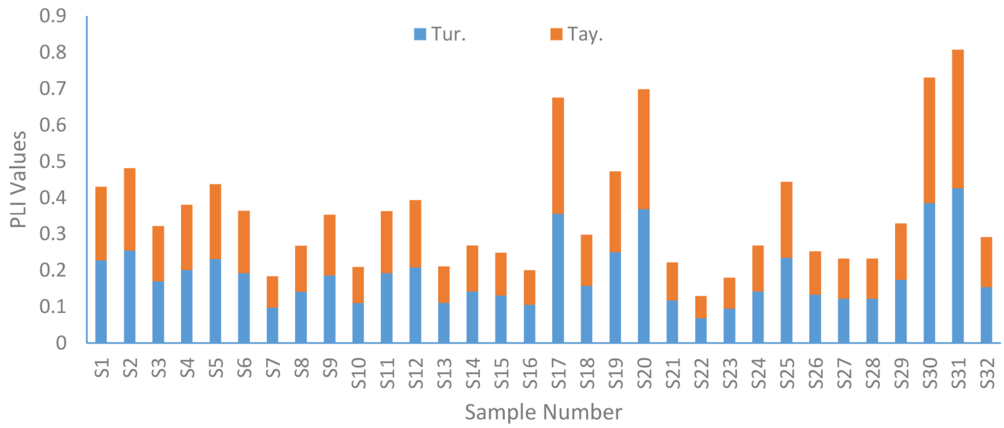

Figure 5. Results of PLI in the study area based on background values [37,38].

Table 4. Distribution of samples in the ranges established by the SQG, according to the PTE concentrations (mg/kg).

Metal	**Mean Concentration**	**Sediment Quality Guideline ***		**% of Samples within Ranges of the Sediment Quality Guideline**		
		ERL	**ERM**	**<ERL**	**>ERL and <ERM**	**>ERM**
Cu	2.44	34	270	100	0	0
Ni	11.76	20.9	51.6	87.5	12.5	0
Pb	2.57	46.7	218	100	0	0
Zn	6.18	150	410	100	0	0
As	2.38	8.2	70	100	0	0
Cr	8.68	81	370	100	0	0
Hg	0.766	0.15	0.71	0	47%	53%

Note(s): * ERL = effects-range—low, ERM = effects-range—median [33].

3.3. Potential Sources of PTEs

Pearson's correlation was conducted to identify the factors and sources of chemical constituents in the studies on environmental pollution [20,49]. The results revealed high positive correlations between many elemental pairs, except Hg (Table 5), implying different behaviors and sources of Hg compared to the remaining PTEs. The high positive correlation between Zn and all the investigated PTEs except Hg suggested that Zn, Cr, Cu, Ni, and Pb might be associated with a natural source; in particular, their average EF were less than 3.00 [50]. Arsenic (As) is severely enriched (average EF = 10–25) and positively correlated with Cu, Ni, and Zn, indicating mixed geogenic and anthropogenic sources [51]. Differently, Hg is negatively and weakly correlated with the remaining PTEs, was severely enriched, and might be derived from anthropogenic factors, mostly from oil pollution, sewage, and industrial effluents spreading nearby Al-Jubail industrial city.

Table 5. The correlation matrix of the analyzed PTEs.

	As	**Cr**	**Cu**	**Ni**	**Pb**	**Zn**	**Hg**
As	1						
Cr	0.495 **	1					
Cu	0.520 **	0.879 **	1				
Ni	0.556 **	0.817 **	0.731 **	1			
Pb	0.363 *	0.550 **	0.551 **	0.571 **	1		
Zn	0.580 **	0.790 **	0.897 **	0.757 **	0.680 **	1	
Hg	−0.050	0.063	0.059	0.278	0.094	0.140	1

Note(s): ** Correlation is significant at the 0.01 level (2-tailed). * Correlation is significant at the 0.05 level (2-tailed).

The principal component analysis indicates the similarity between PTEs and documents their potential pollution sources [52,53]. Herein, PCA divided the PTEs into two components accounting for 61.72% and 15.12% of the total variance (Table 6, Figure 6). The results of PCA strongly concur with those of the Pearson's correlation. As, Cr, Cu, Ni, Pb, and Zn were all highly concentrated in PC1, which primarily suggests the lithogenic sources came from the weathering of rocks with some human contributions for As and Pb [50,54]. PC2 presents a positive loading for Hg, indicating industrial sources may be from petrochemicals of the Al-Jubail industrial city and domestic wastes [46].

Table 6. Principal component loadings and the explained variance for the two components.

	Component	
	1	**2**
As	0.668	−0.289
Cr	0.906	−0.058
Cu	0.916	−0.087
Ni	0.888	0.176
Pb	0.724	0.032
Zn	0.937	−0.008
Hg	0.158	0.966
% of Variance	61.72	15.12
Cumulative %	61.72	76.84

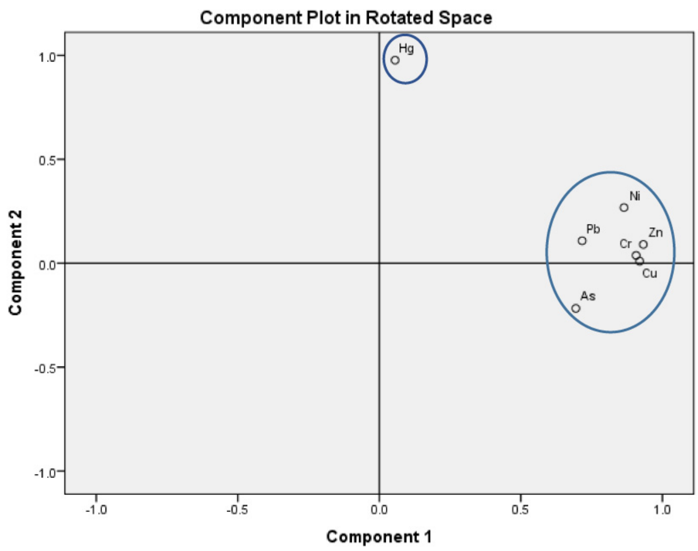

Figure 6. Factor analysis and distribution of PTEs in two component plots.

4. Conclusions

This study highlights the contamination and environmental risk of PTEs in surface marine sediments collected from the coastal area between Al-Jubail and Al-Khafji, Saudi Arabia. Pb, Zn, Ni, Cu, and Cr show no to minor enrichment and minor differences based on the two background references. As and Hg suggest severe enrichment with considerable differences regarding EF and CF. All measurements of the Cu, Pb, Zn, As, and Cr were below the ERL of the SQG, indicating that the studied coastal sediments do not pose a risk to the benthic communities as a result of the presence of these metals. Ni and Hg represent a risk for benthic communities in four and 17 sampled areas, respectively. The m-ERMQ suggests a 9% probability of toxicity for benthic organisms in the studied area. Most of the investigated PTEs indicated natural with some anthropogenic factors, while Hg was mostly derived from anthropogenic effects.

Supplementary Materials: The following supporting information can be downloaded at: https://www.mdpi.com/article/10.3390/w15030573/s1, Table S1: Coordinates of the sediment samples and concentration of the selected PTEs (mg/kg).

Author Contributions: Conceptualization, H.A. and A.S.E.-S.; methodology, H.A. and A.S.E.-S.; writing—original draft preparation, H.A., A.S.E.-S., S.Q. and F.A.; writing—review and editing, H.A., A.S.E.-S., S.Q. and F.A. All authors have read and agreed to the published version of the manuscript.

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