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Distribution and Comprehensive Risk Evaluation of Cr, Cd, Fe, Zn, and Pb from Al Lith Coastal Seawater, Saudi Arabia

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Abstract: Seawater contamination is a global challenge due to its hazardous effects on marine organisms and human health. Twenty-three surface seawater samples were collected from Al Lith intertidal area along the Saudi Red Sea coast to evaluate the ecological risks and document the potential sources of Cr, Cd, Fe, Zn, and Pb. Contamination factor (CF), contamination degree (Cd), water quality index (WQI), and heavy metal pollution index (HPI), as well as multivariate tools were applied. The averages of HMs (µg/L) had the following order: Zn (6.616) > Pb (0.284) > Cd and Cr (0.268) > Fe (0.197). Results of CF showed moderate contamination of seawater with Cd and low contamination for Cr, Fe, Zn, and Pb. However, 26.09% of the samples showed considerable contamination for Cd. Average Cd values revealed low contamination for HMs, while 17.39% of the samples showed moderate contamination. HPI average values indicated medium pollution for Al Lith seawater, meanwhile 13 samples reported high pollution. The higher HPI values were reported in samples characterized by higher concentration of HMs, particularly Cd and Zn. Correlation matrix and principal component analysis suggested anthropogenic sources for Pb and Zn, mostly from industrial and agricultural effluents, landfilling and domestic wastewaters, apart from their natural sources.

Keywords: risk assessment; Zinc; lead; cadmium; seawater; Red Sea

1. Introduction

Coastal areas exhibit remarkable diversity in terms of geology, ecology, biology, and topography. Sea-level rise, climate change, and human activities are factors that lead to alterations in these vulnerable and constantly changing settings [1–3]. In addition, the growth of industries and the fast economic expansion of coastal regions contribute significantly to environmental pollution. These places release millions of tons of pollutants into water bodies [4,5].

Heavy metals (HMs) enter to the aquatic ecosystem due to natural processes and anthropogenic activities including weathering of nearby rocks weathering, atmosphere deposition, agricultural, industrial, storm water, sewage treatment, and domestic wastes. The arsenic, mercury, chromium, cadmium, lead, nickel, copper, and zinc are the eight most common types of coastal contaminants listed by the Environment Protection Agency [6]. Some HMs, such as As, Pb, Cr, Cr, and Ni are characterized by their persistence, bioaccumulate in aquatic organisms, and their influence varies depending on their concentration and available chemical forms [7,8]. Additionally, toxic HMs can be taken up by marine organisms, entering the food chain and being potentially transferred to higher trophic levels and then affecting human health [8,9]. The concentration of any HMs in humans can be too low, sufficient, or harmful depending on the levels of these HMs in the environment and the extent of exposure [10].

The consumption of marine species that are contaminated with potentially harmful HMs, such as lead, arsenic, cadmium, and nickel, is the primary route by which humans are exposed to these metals. HMs pose a threat not only to human health but also to the biodiversity of aquatic life, as demonstrated by the ongoing decrease in the population and variety of freshwater fish and other aquatic species. The human body experiences a range of chronic health issues as a result of metal toxicity. These include brain and nerve disorders, blood disorders, structural damage, alterations in kidney function, dermal lesions, and various types of cancer such as skin, lung, bladder, and kidney cancer. Additionally, metal toxicity can lead to skin changes like hyperkeratosis and pigmentation changes, as well as mineralization of bones and teeth. Other symptoms may include stomach irritation, vomiting, and diarrhea [8,11,12].

The Saudi Red Sea coast along Jeddah, Yanbu, Duba, Sharma, Al-Wajh, Jazan, and Sharm Al-Kharrar has been intensively studied regarding HM contamination in sediments and seawaters [e.g., 13-28]. Al Lith area is characterized by economic growth and the expansion of the population in the last decade. Moreover, the coastal area receives flooding, industrial, and domestic discharges which uploaded with trace elements. Finally, no study has been conducted on the seawater monitoring in Al Lith coastal area. Therefore, the main purposes of the present study are to assess the ecological risk assessment of Cr, Cd, Fe, Zn, and Pb in Al Lith seawater, Red Sea coast, Saudi Arabia using CF, Cd, and HPI; and to identify the possible sources of HMs utilizing statistical analysis.

2. Materials and Methods

2.1. Study Area and Sampling

Al-Lith City is located in the Makkah region of Saudi Arabia, approximately 180–200Km to the south of Jeddah City. The basement rocks of the Arabian Shield are found in the eastern portion of the Al Lith area and run parallel to the Red Sea. On the other hand, the western half of the area is covered with deposits of sand, gravel, silt, and mud from the Quaternary period [29,30]. A total of twenty-three samples of saltwater from the subtidal zone were obtained (Figure 1). The sediment in the beach under study consists of mud, fine to coarse sands, and terrestrial gravels. It contains various marine organisms such as corals, gastropods, bivalves, echinoids, seagrass, and foraminifers. These organisms have been transported to the beach by waves and currents [31–33].

At each site, several samples were taken using 500-ml clean, sterilized polyethylene bottles. Prior to sampling, the bottles were washed with seawater at the same site three times [24,25]. The selection of sample locations was decided using GPS technology. To prevent precipitation and sorption to the container walls, the samples were acidified with 5 mL of 10% HNO3 [34]. Graphite furnace atomic absorption spectroscopy (Ananlytica Jena) was used to measure the concentrations of Cr, Cd, Fe, Zn, and Pb in Yarmouk University, Jardon.

Prior to measurements, the saltwater samples were diluted by a factor of 10 with deionized water. Single element and multi-element standard solutions (Merck, Sigma-Aldrich) were used to prepare all multi-element stock solutions. The single and multi-element working standards were generated by sequentially diluting the stock solutions using volume/volume dilution. The calibration standards and blank solution were acid-matched with the sample solutions using a 1% (v/v) HNO_3 and 1% (v/v) HCl solution. The sea water samples were filtered using Whatman filter paper, if necessary, and were acidified immediately after filtration. The calibration levels for each element were selected based on the prescribed threshold values. A minimum of five calibration standards were utilized for each element, as outlined below. Initially, the samples were examined using Flame Atomic Absorption Spectroscopy (FAAS) to identify any outliers. Subsequently, if necessary, the samples were re-analyzed using Graphite Furnace Atomic Absorption Spectroscopy (GFAAS) to ensure the accuracy of the results. The internal standard quality control samples were evaluated and showed a strong connection with the estimated values. Duplication was employed to calculate the sample mean reading (Table S.1).

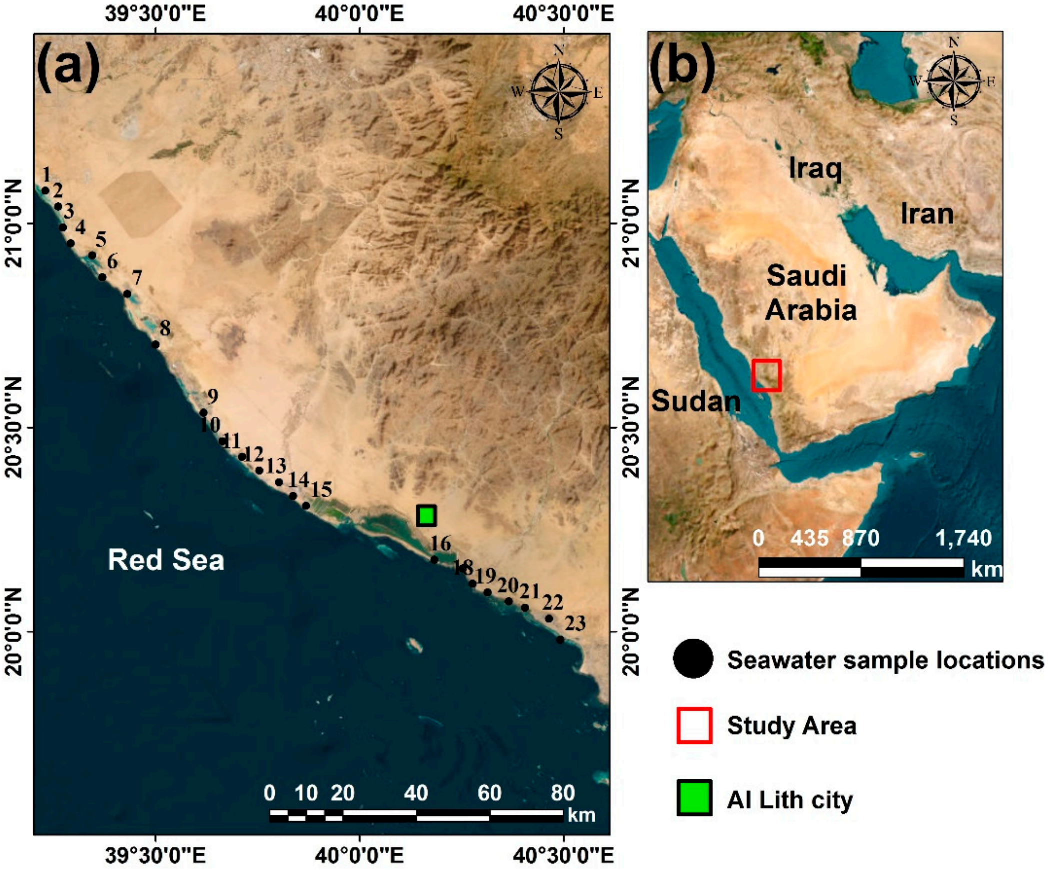

Figure 1. Location map and sampling sites of Al Lith seawater.

2.2. Pollution Indices and Multivariate Analyses

Correlation analysis (CA) and principal component analysis (PCA) are widely used statistical methods for identifying the origins of HM contamination and interpreting datasets [35,36]. The contamination factor (CF), contamination degree (Cd), modified contamination factor (mCd), and heavy metal pollution index (HPI) were utilized to evaluate the level of contamination in seawater. The modified contamination factor (mCd) can serve as a water quality indicator (WQI) for a thorough evaluation of saltwater contamination and quality [37,38]. The calculation methods of the contamination indices utilized in this investigation are summarized in Equations 1-5 and Table 1 [39,40].

$$\text{CF} = C\ (\text{HMs})/C\ (Background) \quad (1)$$

$$\text{Cd} = \sum CF \quad (2)$$

$$WQI = \text{mC}_{\text{deg}} = \sum CF/\text{n} \quad (3)$$

$$\text{HPI} = \frac{\sum WiQi}{\sum Wi} \quad (4)$$

$$\text{Q}_{\text{i}} = \sum(\frac{[Mi(-)Ii]}{Si-Ii}) \quad (5)$$

CF is the contamination factor, C (HMs) is the analytical value and C ($Background$) is the upper permissible concentration for the ith component, Qi is the sub-index of the ith parameter, Wi is the unit weight for the ith parameter, Mi, Ii and Si are the monitored HM, ideal and standard values of the ith parameter, respectively. The sign (-) indicates numerical difference of the two values, ignoring the algebraic sign.

Table 1. Classification of the contamination indices applied in this study [39–41].

Index	Value	Contamination level
CF	CF < 1	Low contamination factor
	1 ≤ CF <3	Moderate contamination factor
	3 ≤ CF < 6	Considerable contamination factor
	CF ≥ 6	Very high contamination factor
C_d	$C_d < 4$	Low contamination degree
	$C_d = 4\text{-}8$	Moderate contamination degree
	$C_d > 8$	Very high contamination degree
mC_d	< 1.5	Uncontaminated
	$1.5 \leq mC_d < 2$	Slightly contaminated
	$2 \leq mC_d < 4$	Moderately contaminated
	$4 \leq mC_d < 8$	Moderately to heavily contaminated
	$8 \leq mC_d < 16$	Heavily contaminated
	$16 \leq mC_d < 32$	Severely contaminated
	$mC_d \geq 32$	Extremely contaminated
HPI	HPI < 5	Low pollution
	HPI = 5–10	Medium pollution
	HPI > 10	High pollution

3. Results and Discussion

3.1. Distribution and Contamination Assessment of HMs

Supplementary Table S2 displays the lowest, highest, and average levels of HMs, as well as the outcomes of the pollution indices employed in this study. The average HMs took the following descending order: Zn (6.616 μg/L) > Cd (0.59 μg/L) > Pb (0.284 μg/L) > Cr (0.26860 μg/L) > Fe (0.197 μg/L). The distribution of HMs in each sample location within the research area exhibited a fluctuating pattern without a specific orientation (Figure 2). Nevertheless, certain individual samples exhibited variations in HM amounts, either increasing or decreasing. For example, sample 1 located in the northern region of the research area has the greatest concentrations of Zinc and Lead, measuring 30.800 and 1.053 μg/L, respectively. Conversely, samples 20 and 18 in the southern region display the lowest levels of these two HMs, measuring 1.00 and 0.021 μg/L, respectively. The samples with the highest concentrations of chromium and iron were 20 and 23, respectively, in the southern region of the research area. The concentration of chromium in sample 20 was 0.423 μg/L, while the concentration of iron in sample 23 was 0.414 μg/L. On the other hand, the samples with the lowest concentrations of chromium and iron were 19 and 2, respectively. The concentration of chromium in sample 19 was 0.018 μg/L, while the concentration of iron in sample 2 was 0.021 μg/L. In relation to Cd, samples 2 and 4 in the northern region of the research area exhibited the greatest and lowest concentrations (1.020 and 0.100 μg/L, respectively). The observed rise in HM values in certain individual samples can be ascribed to the composition of the mud and fine sediment present at these locations, which possess the capacity to retain HMs and then release them into the water column [42].

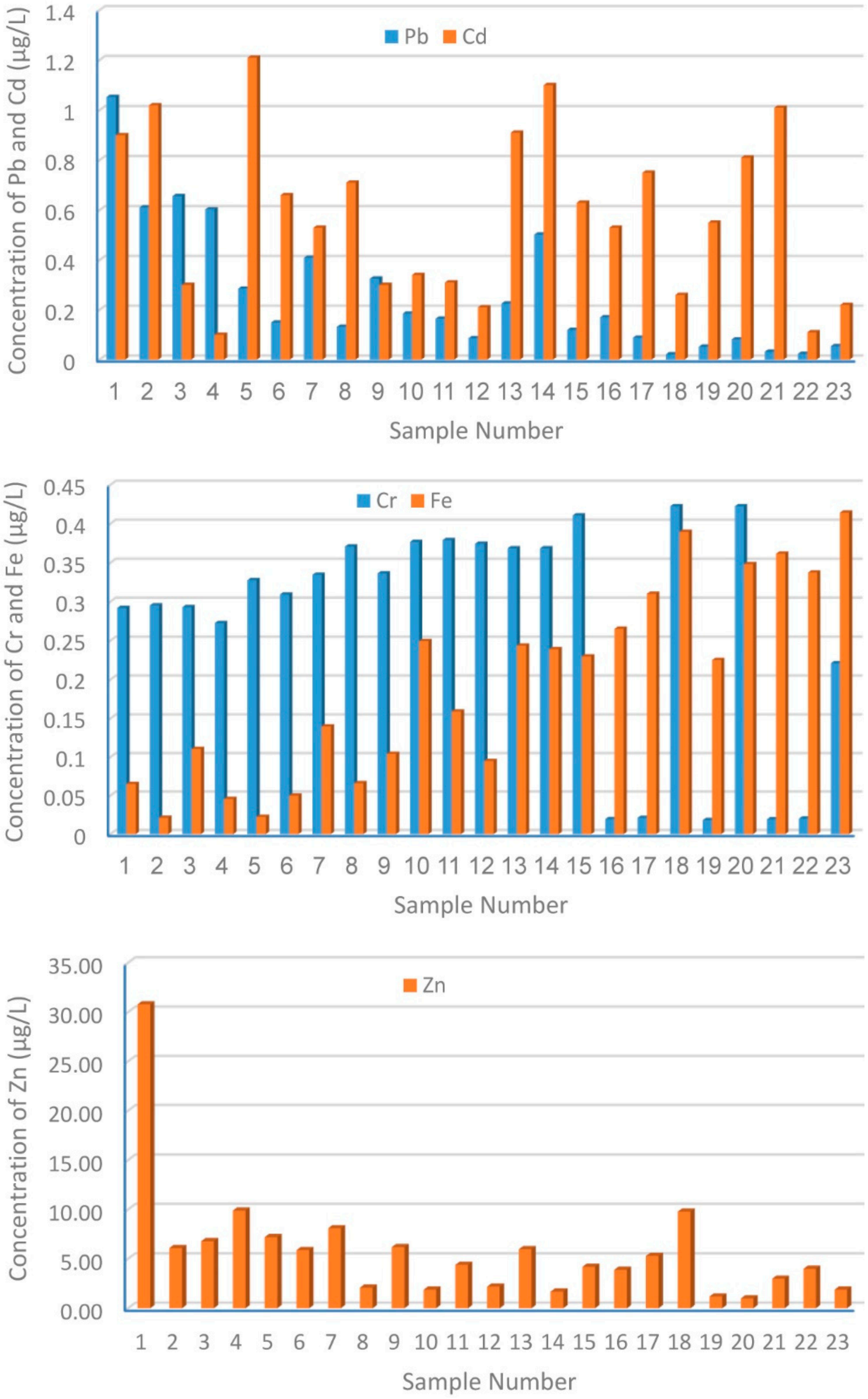

Figure 2. Distribution of heavy metals per sample locations in Al Lith seawater.

The average levels of HMs in Table 2 were less than the maximum admissible concentration [43]. Average Cr and Fe values were less than those reported from Al-Uqair coastline, Saudi Arabia [44], Gulf of Aqaba, Saudi Arabia [45], Yanbu coastline, Red Sea, Saudi Arabia [46], Al-Khobar, Arabian Gulf, Saudi Arabia [47], and Tarut Island, Saudi Arabia [48]. Moreover, average Zn value was lower than those reported from Al-Uqair and Al-Khobar seawaters, Arabian Gulf, Saudi Arabia [44,47]. Differently, average Zn and Cd values were greater than those reported from Yanbu, Jazan coastlines, Red Sea, Sharm Al-Kharrar lagoon Saudi Arabia, and Gulf of Aqaba [20,24,46,49].

Table 2. Average concentration of HMs (μg/L) in Al Lith seawaters as compared to worldwide and maximum admissible concentrations.

References	Cr	Fe	Zn	Pb	Cd
Present study	0.268	0.197	6.616	0.284	0.59
Jazan coastline, Saudi Arabia [20]	1.28	-	1.04	2.27	0.06
Sharm Al-Kharrar lagoon, Saudi Arabia [24]	0.26	-	4.19	0.28	0.06
Red Sea-Gulf of Aqaba, Saudi Arabia [25]	0.26	1.81	5.51	1.31	0.05
Yanbu coastline, Red Sea, Saudi Arabia [28]	0.72	0.983	1.67	0.28	0.31
Maximum admissible concentration [43]	50.0	200	40.0	10.0	3.00
Al-Uqair coastline, Saudi Arabia [44]	9.64	6.13	6.72	0.26	0.05
Gulf of Aqaba, Saudi Arabia [45]	0.96	15.255	3.32	0.20	0.03
Al-Khobar, Arabian Gulf, Saudi Arabia [47]	1.38	3.54	16.21	0.04	0.11
Tarut Island, Saudi Arabia [48]	12.95	6.52	0.97	0.48	0.03
Gulf of Aqaba [49]	-	1.78	0.24	0.32	0.57
Al-Khafji, Arabian Gulf, Saudi Arabia [50]	0.70	0.99	1.53	0.28	0.07
Rosetta coast, Mediterranean Sea, Egypt [51]	0.004	0.22	0.013	0.006	-

3.2. Ecological Assessment of HMs

The evaluation of the contamination factor (CF) at different sampling sites in Al Lith seawater demonstrated a wide range of circumstances (Table S.3). Overall, the seawater samples showed varying levels of contamination for different HMs. The CF values for the HMs followed the order: Cd (1.97) > Zn (0.331) > Pb (0.284) > Cr (0.054) > Fe (0.00099). This indicates that Cd had a moderate contamination factor, while the other HMs had relatively low levels of contamination [38]. Samples 1, 2, 5, 13, 14, and 31, which account for 26.09% of the samples, had CF values more than 3 for Cd, indicating a significant contamination factor (Table S.2). Furthermore, Sample 1 exhibited a moderate contamination factor for Pb and Zn, with values of 1.053 and 1.54, respectively.

The contamination degree (C_d) values exhibited substantial diversity among the many investigated sites along the Al Lith shoreline. The C_d results varied from 0.60 to 5.65, with an average of 2.61, suggesting that the overall examined seawaters had low contamination levels [39,52]. Nevertheless, the analysis of C_d values for each sample location (Figure 3) revealed that samples 1, 2, 5, and 14 (which account for 17.39% of the total samples) exhibited C_d levels ranging from 4 to 8, indicating a moderate level of contamination [53]. The distribution pattern of the mCd (WQI) closely resembles that of Cd. The values varied from 0.119 to 1.130, with an average of 0.521, suggesting that the saltwater was not polluted [40].

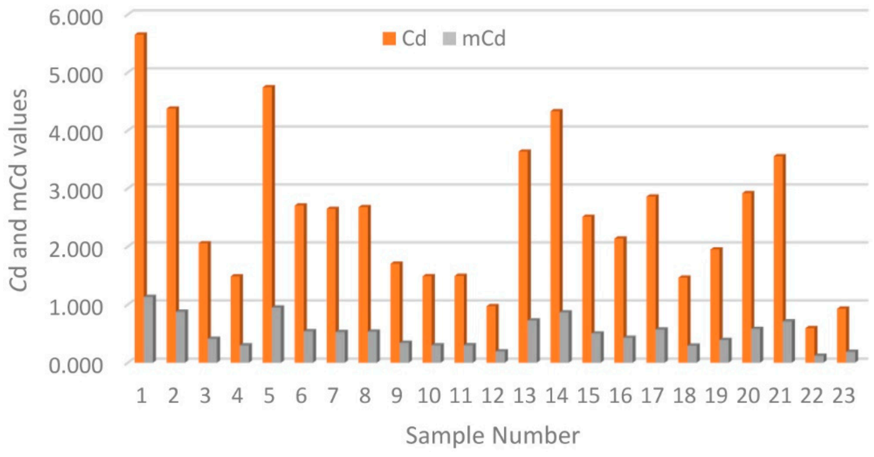

Figure 3. Distribution of Cd and mCd per sample locations in Al Lith seawater.

Results of heavy metal contamination index for Al Lith seawater ranged from 3.10 to 29.37, with an average of 14.90 (Figure 4, Table S. 1), indicating a moderate level of pollution [28]. Out of the total

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samples, 13 (1, 2, 5-8, 13-17, and 19-21) exhibited HPI values exceeding 10, indicating a high level of pollution. Additionally, 7 samples (3, 9-12, 18, and 23) had HPI values ranging from 5 to 10, suggesting a moderate level of pollution. The remaining 3 samples were classified as having low pollution [41,54]. The samples with higher concentrations of HMs, including Cd, Pb, and Zn, revealed higher HPI values. The sweater samples with higher pollution indices were primarily taken at the mouth of Wadi Al-Lith. This suggests that the related HMs may have originated from the volcanic and metamorphic rocks of the Arabian Shield [20].

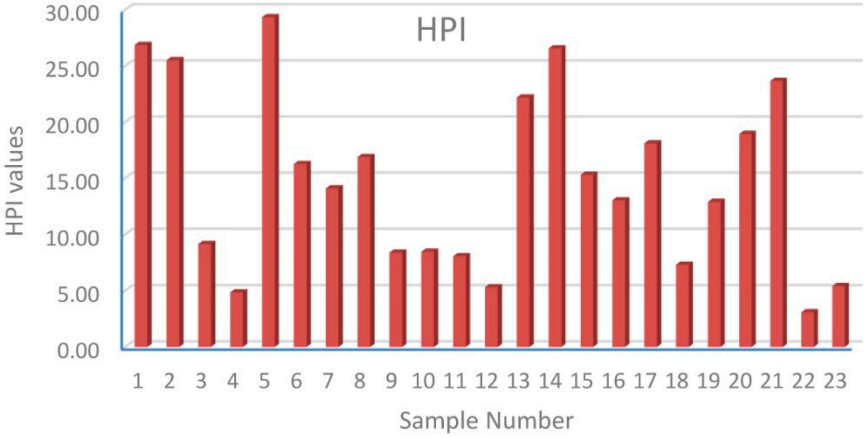

Figure 4. Distribution of HPI per sample locations in Al Lith seawater.

3.3. *Potential Sources of HMs*

Industrial operations and the use of sewage sludge or superphosphate have significantly contributed to the introduction of lead (Pb) and cadmium (Cd) into the environment [8,10]. Lead-acid batteries are the primary global application of Pb. Furthermore, Cd is mostly utilized in the manufacturing of Ni-Cd and Ag-Cd batteries. Additionally, the nonferric metal industry and agricultural practices are human-made sources of zinc (Zn). The bauxite parent material naturally contains a high concentration of zinc.

Correlation analysis is a method used to quantify the degree of linear relationship between two variables and to find the sources of HMs that have a strong correlation [55,56]. The correlation matrix in Table 3 revealed negative and weak correlations among all HM pairs, except for a positive correlation between Pb and Zn (r = .738). This suggests that the sources of Pb and Zn in the study area are primarily anthropogenic, originating from industrial and agricultural effluents, landfilling, and domestic wastewaters, in addition to their natural sources [57,58]. The absence of substantial correlations among numerous HM pairs suggests the presence of multiple sources of contamination [59,60].

Table 3. Correlation matrix of the investigated HMs.

	Cr	**Cd**	**Fe**	**Zn**	**Pb**
Cr	1				
Cd	-.175	1			
Fe	-.326	.228	1		
Zn	.099	-.019	-.347	1	
Pb	.214	-.176	-.596**	**.738****	1

**. Correlation is significant at the 0.01 level (2-tailed).

Principal component analysis (PCA) categorizes HMs into many main components by examining the interplay among various variables [60]. PCA in the study area produces two PCs, which account for 46.10% and 22.41% of the variance, respectively. Together, these components explain a total of 68.51% of the variation (Table 4). The PC1 shows a substantial positive correlation with Zn (0.757) and Pb (0.903), but the second PC2 has a positive correlation with Cd (0.681), indicating distinct anthropogenic origins [61–63].

Table 4. Principal component loadings and variance percentage for the extracted two components.

	Component	
	PC1	**PC2**
Cr	.445	-.568
Cd	-.329	**.681**
Fe	-.782	.159
Zn	**.757**	.497
Pb	**.903**	.250
% of Variance	46.10	22.41
Cumulative %	46.10	68.51

4. Conclusions

This study employed the use of CF, Cd, mCd, WQI, and HPI, as well as CM and PCA to evaluate the ecological hazards posed by HMs in seawaters along the Al Lith coastal area and identify the possible origins of these HMs. The subsequent discoveries were acquired:

1. The HMs were arranged in descending order according to their average. Zinc (6.616 µg/L) has a higher concentration than Cadmium (0.59 µg/L), which has a higher concentration than Lead (0.284 µg/L), which has a higher concentration than Chromium (0.26860 µg/L), which has a higher concentration than Iron (0.197 µg/L). The distribution of HMs per sample location in the research area exhibited a shifting pattern, with certain individual samples displaying varying levels of rise or reduction. The average levels of HMs were below the maximum allowable concentration set by the World Health Organization.

2. The studied seawater showed low to moderate contamination levels based on the measurements of CF, Cd, mCd, WQI, and HPI. Nevertheless, certain individual samples exhibited significant contamination and elevated pollution levels. The samples with higher concentrations of heavy metals, including cadmium (Cd) and zinc (Zn), showed higher levels of contamination.

3. Both CA and PCA proposed that the basement rocks of the Arabian shield are natural sources of HMs, but anthropogenic sources are also responsible. Nevertheless, human activities have been identified as the main causes of Pb, Cd, and Zn contamination, mostly through the release of pollutants from industrial, agricultural, and home sources.

Supplementary Materials: The following supporting information can be downloaded at the website of this paper posted on Preprints.org, Table S1: The internal standard quality control samples; Table S2: Concentration of HMs (µg/L) along with results of some contamination indices applied in this study; Table S3: The Contamination factor (CF) for heavy metal(loid)s in seawater.

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Conflicts of Interest: “The authors declare no conflicts of interest”.

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