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Contamination Evaluation of Heavy Metals in a Sediment Core from the Al-Salam Lagoon, Jeddah Coast, Saudi Arabia

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Abstract: The Al-Salam Lagoon is one of the recreational sites along the Jeddah coast, showing the environmental impacts of urbanization along the coast. A sediment core (220 cm) was collected from the intertidal zone to evaluate the heavy metals (Fe, Mn, Cr, Ni, Cu, Zn, and Pb) and geochemical indices (contamination factor, geo-accumulation index, and pollution load index). In the organic-rich muddy sediments (0–100 cm), there is a high metals content and a pollution load index of ~3, indicting anthropogenic impacts with high Cu contamination (CF:12) and moderate Fe, Mn, Cr, Ni, Zn, and Pb contamination (CF: <3). The organic matter and heavy metals washed through surface run-off from the land and deposited as urban waste. Down the core, consistent metals concentration, CF, and I_{geo} trends indicate a common pollutant source and pollution load variations over time. In the sediment section (70–40 cm), a high organic matter, metal concentration, CF, I_{geo}, and PLI value ($\geq$5) suggest an uncontrolled pollution load. The decreased and stable trends of environmental indicators toward surface sediments suggest measures taken to control the pollution along the Jeddah coast. Below 110 cm, the carbonate-rich sediments have low organic matter and metals, showing an unpolluted depositional environment. The negative geo-accumulation index implies a geogenic source and indicates no anthropogenic impacts as inferred from low (~1.0) CF and PLI.

Keywords: Al-Salam lagoon; contamination assessment; heavy metals; LOI; Jeddah coast

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1. Introduction

The coastal areas are the repository of the waste material generated by multiple human activities. The effluents, discharged from the land, deposit heavy metals in coastal sediments [1]. In the aquatic environment, adsorbed heavy metals flocculate, co-precipitate, and deposit onto the sediments [2,3]. Heavy metals pose a threat to the ecosystem [4]. The sediments' geochemistry provides information about environmental pollution [3,5–8]. The surface sediment record explains contamination levels in the present, whereas sediment cores provide a historical record of variations in natural and anthropogenic input [9–12].

Jeddah is a fast-developing port city in the Kingdom of Saudi Arabia. The impacts of urbanization along the Jeddah coast have threatened the coastal system. The harbor infrastructures, urbanized centers, industrial zones, and sewage outfalls are the important sources of contaminants in the sediments depositing along the Jeddah coast. Studies reported pollution and its adverse impacts along the Jeddah coast [3,13–21]. Al-Salam Lagoon is a recreational area of the Jeddah coast and appears to be stagnant water with a thick scum of blue-green algae. Risk et al. [22] found high concentrations of Ni, Cu, and Zn in the lagoon sediments and categorized them as heavily polluted. South of the Al-Salam Lagoon, the Al-Arabian Lagoon shows high nitrogen, phosphorus, trace elements, organic matter, and hydrocarbons in the waters and sediments [14,17,23]. Published environmental records, especially the geochemical aspects of the sediments, from Al-Salam

Lagoon are lacking. This research evaluates the accumulation of metals (Fe, Mn, Cr, Zn, Ni, Cu, and Pb) in a 220 cm long sediment core (SALAM1) collected from the Al-Salam Lagoon (21°31′55.9″ N, 39°09′25.2″ E). The aims were (i) to determine the distribution of organic matter, which was determined by Loss on ignition (LOI), carbonate, mud, sand contents, major elements such as Fe and Mn, and heavy metals such as Cr, Zn, Ni, Cu, and Pb, in the sediments along the core and (ii) to evaluate the severity of the contaminants using geochemical environmental indicators: the contamination factor (CF), the geo-accumulation index (I_{geo}), and the pollution load index (PLI). This may help evaluate the evolutionary trends of anthropogenic impacts along the Jeddah coastal zone and the extent of environmental pollution in the Al-Salam Lagoon since the developmental activities in the area. The best understandings of the evolutionary trends of anthropogenic impacts and the conditions that existed prior to the industrial revolution may help decision-makers to form possible solutions for rehabilitation such as this lagoon. Therefore, this study also provides new information about the historical evaluation of heavy metals in the area through the late Holocene. In addition, it provides a typical case study that can be helpful for any investigation on the other lagoons worldwide.

2. Materials and Methods

2.1. Study Area

The Al-Salam lagoon is a recreational site in the central part of the Jeddah coast (Figure 1). Since the 1970s, a large amount of sewage wastewater has been discharged in the Jeddah coastal waters. El-Syed [20] reported that the Al-Arbaeen and Al-Shabab lagoons, located south of the study area, receive about 100,000 m^3 of sewage water daily. The sewerage discharge has converted the lagoons into polluted lagoons. The sewage discharge resulted in high nutrients, biochemical oxygen demand (BOD), chemical oxygen demand (COD), fecal bacteria, high phosphate, high ammonia, and nitrate [22]. In the Arbaeen and Al-Shabab lagoons, the sediments show heavy metal enrichment and nutrients [14]. In comparison, the Al-Salam lagoon surface sediments are categorized as polluted lagoon by high concentrations of Ni, Cu, and Zn [22]. The lagoon appears to be stagnant water, showing a thick scum of blue-green algae as a result of eutrophication. North-northwestern winds move water toward the open sea. These environmental conditions increase metal pollution in the sediments and pose ecological threats in the coastal area, especially for public health.

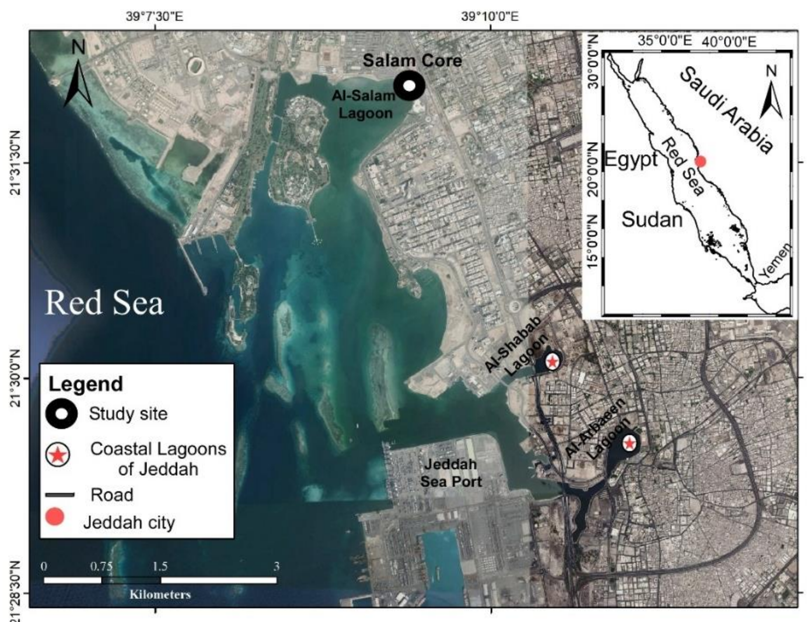

Figure 1. Location map of sediment core Salam1 and coastal lagoons of the Jeddah city, Saudi Arabia.

2.2. Sediment Sampling and Analysis

The sediment core Salam1 (21°31′55.9″ N, 39°09′25.2″ E) was collected from 0.5 m water depth of the Al-Salam Lagoon (Figure 1). Core sediments were sub-sampled at 3 and 5 cm intervals in the upper section (0–100 cm; 34 samples) and the lower section (100–220 cm; 24 samples) of the core, respectively. These differences in sampling resolution were based on lithology changes. The wet sieving method [24] was used for grain size analysis. The sediment fraction retained on the sieve's 2 and 0.063 mm mesh diameter were dried and weighed to calculate the percentage of >2 mm (gravel), 2–0.063 mm (sand), and <0.063 mm (mud). Organic matter was determined by the (LOI) method [25]. A total of 1 g of dry ground sediments was ignited at 550 °C for 4 h. The ash was re-weighed to determine the total weight loss as LOI percent. Calcium carbonate ($CaCO_3$) content was determined by the gasometric technique using a Calcimeter [26]. For heavy metal analysis, sediment samples were digested with Aqua Regia. The undigested portion filtered off through a Whatman 540. A clear solution was diluted to 50 mL with distilled water and used for geochemical analysis using Atomic Absorption Spectrophotometry (AAS) Perkin Elmer (A Analyst 800). For quality assurance, analytical blanks were run, and the concentrations were determined using standard solutions. Each sample was analyzed in triplicate, and an average result was used in this study. The precision of heavy metals has been expressed as a relative standard deviation (RSD). It was 5.09% and 1.02% for iron and manganese, whereas the accuracies of Cr, Zn, Cu, Ni, and Pb were 1.72%, 1.63%, 0.72%, and 2.40%, respectively.

3. Results

3.1. Core Lithology and Sediment Texture

The sedimentary record of the Salam1 core consists of three distinct intervals (Figure 2a). The basal interval (220–110) is comprised of white to grey bioclastic sandy mud. It contains ≤10% gravel. The coarse sediments (40% sand with 10% gravel) contain abundant skeletal remains. The middle interval (100–65) is composed of white to grey mud with faint laminations. It is sharply contacted with the lower interval, and it is dominated by >30% of mud and 60% of sand fractions. The upper interval (65–0) represents the top of the core and consists of black mud devoid of shells and sell fragments. This black mud is composed mainly of black sludge and slurry with remains of plastic, indicating anthropogenic activities during this interval. The sediment texture of this interval is muddy (60–90% mud) with sandy intercalations between 20 and 40 cm (Figure 2b,d). Overall, the Salam 1 core may be correlated with the core of the Shuaiba Lagoon coast south of Jeddah [27], which is dated as of the late Holocene (last 3.6 ka).

3.2. Calcium Carbonate and Organic Matter

Carbonate ($CaCO_3$) content in the sediments ranges from 30.59% to 61.76% (Supplementary Table S1). The carbonate profile (Figure 2e) shows low (<40%) $CaCO_3$ in the muddy sediments. $CaCO_3$ increases to ~60% as the sand increases. Between 70 and 110 cm depth, muddy sediments show high (~60%) $CaCO_3$. In sandy sediments (110 to 220 cm), carbonate fluctuates between 50% and ~60%.

The approximated organic matter in the sediments ranges from 2.2% to 26.71% (Supplementary Table S1). LOI profile (Figure 2f) follows mud variations. A high (5–15%) LOI is noted in the top 70 cm muddy sediments. Calcareous mud (70 and 110 cm) and sandy sediments (110 to 220 cm) show low (≤5%) LOI. Organic matter (av. 6.0%) in the core SALAM1 sediments is higher than the unpolluted Red Sea sediments. Basaham and El-Shater [28] showed low organic matter (0.06–0.45%) for the Red Sea sediments. The vertical distribution of the mud, organic carbon (LOI), and metals (Figure 3a–i) confirms high metal content in the polluted muddy sediments from the Jeddah coast [14].

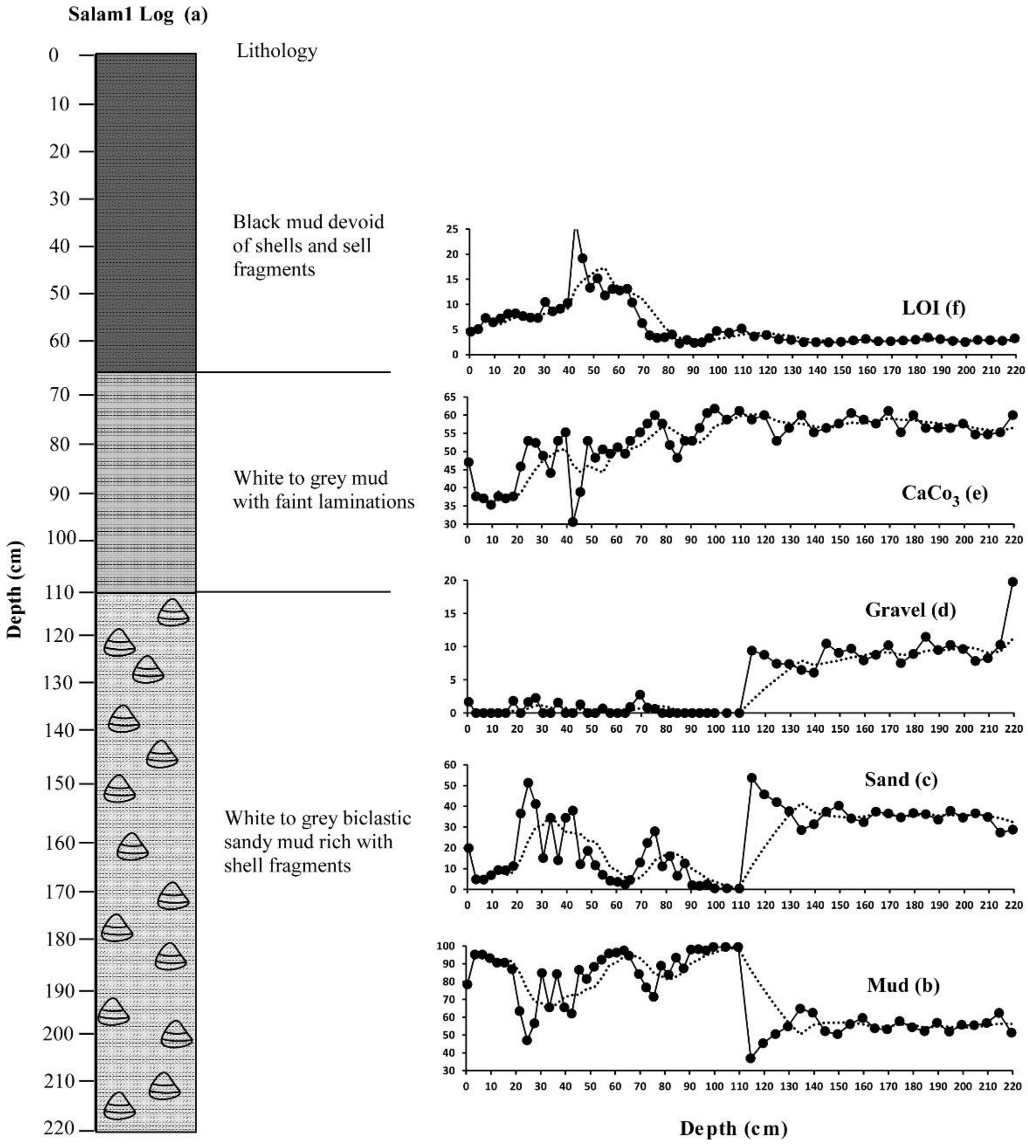

Figure 2. (**a**) Lithologic of the Salam1 sedimentary record, (**b**–**f**) Mud, sand, gravel, LOI%, and $CaCO_3$% distribution in the Salam1 core from Al-Salam Lagoon, the Jeddah coast, Saudi Arabia. (Trend line of 5 average is shown by dots).

3.3. Metals

The heavy metals (Supplementary Table S1) show the following order of their abundances: Zn > Fe > Mn > Cu > Pb > Cr > Ni. The average metal content is higher in the upper half (0–110 cm) than in the lower half (110–220 cm) of the core (Table 1). The average metal concentration is comparable to the published regional and local studies (Table 1). The vertical distribution of the metals displays a progressive upward increase in the concentration level following the mud and organic carbon (LOI) variations (Figure 3a–i). The lower part (110–220 cm) of the core shows low values and uniform distribution.

High Fe and Mn concentration ranges between 350–645 and 300–500 μg g^{-1}, respectively, occurs in the top 30 cm surface sediments (Figure 3c,d). Downward, below 70 cm depth, Fe and Mn content decreases to ≤100 μg g g^{-1}, following the LOI profile, and is uniformly distributed in the bottom core (110–220 cm).

Cr concentrations vary between 32.6 and 109.8 μg g^{-1} with a mean value of 61.0 μg g^{-1} (Supplementary Table S1). The vertical distribution of Cr in the SALAM1 core (Figure 3e) is similar to Fe and Mn. Zinc (Zn) content in sediments of the Al-Salam Lagoon is the highest (830 μg g^{-1}) of the investigated metals. The Zn concentration ranges from 21 to 830 μg g^{-1}, with a mean value of 227 μg g^{-1} (Supplementary Table S1). A trend of vertical distribution (Figure 3f) is similar to other metals. The Zn content in the top 30 cm surface sediments is ≥400 μg g^{-1}. The concentration, between 40 and 70 cm core depths, is relatively high and varies between 200 and 500 μg g^{-1}. Below 70 cm, the Zn content decreases to ≤100 μg g^{-1} and is consistently low ≤ 200 μg g^{-1} in the lower part of the core.

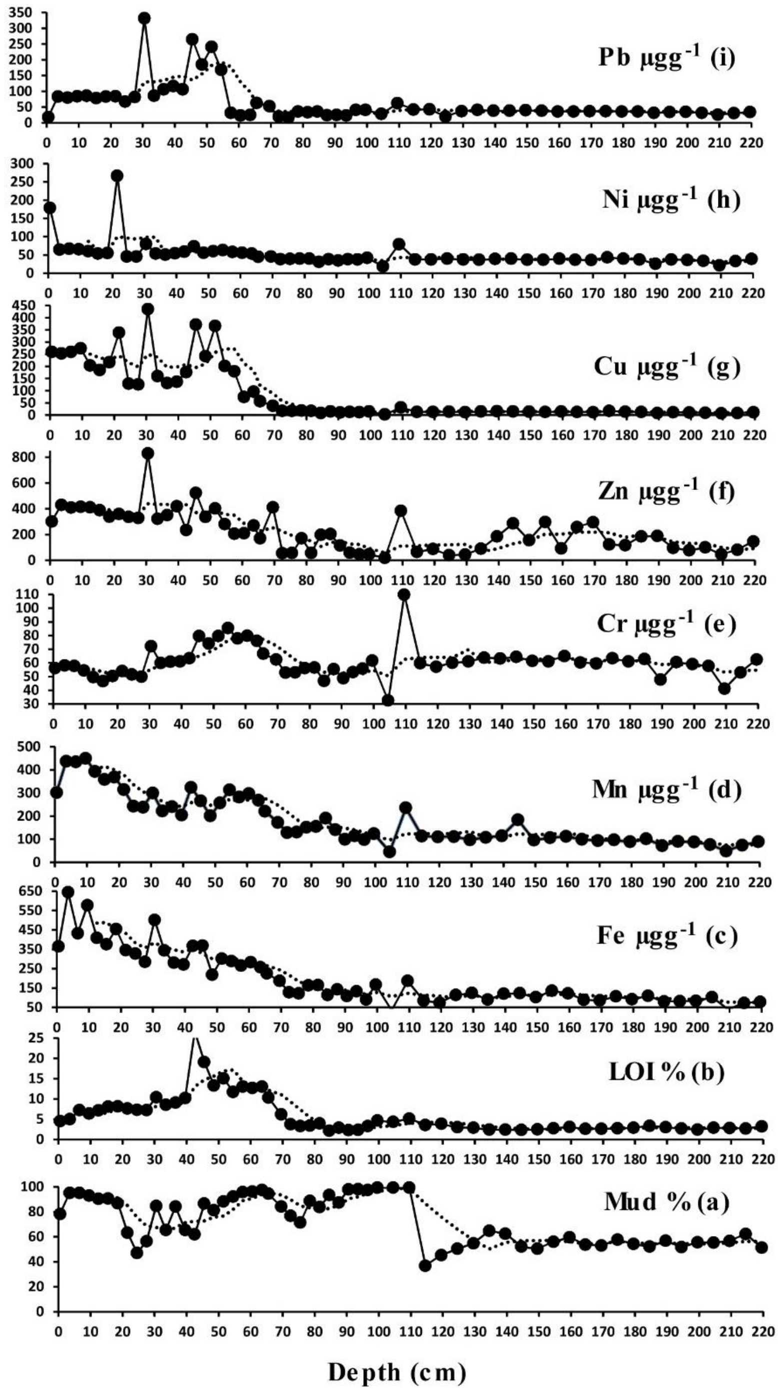

Figure 3. (**a**,**b**)Vertical distribution of mud and LOI %, (**c**–**i**) Vertical distribution of metal concentrations in the Salam1 core from Al-Salam Lagoon along the Jeddah coast, Saudi Arabia (trend line of 5 Average is shown by dots).

Table 1. Comparison of average concentrations (μg g^{-1}) of heavy metals in the core Salam1 sediments from Al-Salam Lagoon, Jeddah coast, and similar published studies from the Red Sea and other world regions.

Location on the Red Sea	Cr	Cu	Fe	Mn	Ni	Zn	Pb	References
				μg g^{-1}				
Al-Salam Lagoon Core Jeddah Total Average (0–220 cm)	60	92	208	188	51	227	64	Present Study
Average of Top section (0–110 cm)	61	141	276	242	60	281	81	
Average of Bottom section(110–220 cm)	59	12	97	99	37	140	35	
Downtown Core Jeddah (0–50 cm)	265	352		678	-	747	382	[29]
Jizan	5.64	16.39	-	9.58	14.32	24.74	3.86	[30]
Sharm Obhur, Jeddah.	144	47	-	674	57	82	5	[31]
Al-Arbaeen Lagoon Jeddah	60	58	-	139	48	118	-	[14]
Al-Shabab Lagoon Jeddah	89	72	-	247	85	234	-	
Jeddah Coast	-	82	-	-	-	179	69	[15]
Jeddah Coast	12–23	17–24	2032–2671	34–205	67–85	52–76	80–99	[16]
Al-Shuaiba Lagoon	39	31	1600	170	13	29	-	[32]
Al-Mejarma Lagoon	58	33.0	1700	303	11	35	-	
Al-Kharar Lagoon	69	44	3180	711	39	55	-	
Al-Kumrah, Jeddah	-	14	19.21	23.87	-	1.01	39.32	[13]
Al-Shoaibah, Jeddah	-	0.5	6.70	3.30	-	0.257	3.06	
Al-Shabab Lagoon Jeddah.	-	163	-	-	-	179	116	[17]
Al-Hoedidah coast Yemen	6–38–38.46	36.70–79.90	7.10–116.4	9.17–24.21	99.67–199.76	4.02–18.25	4.99–6.26	[33]
Gulf of Aqaba								
Jordan	1.12	0.03	-	-	0.43	0.42	4.07	[11]
Persian Gulf Iran	5.7–52.4	1.9–304	-	50–466	-	5–123	1–14	[34]
Mediterranean Sea								
Bizerte lagoon, Tunisia	-	2.81	513.58	71.31	-	33.54	31.61	[9]
Atlantic Coast								
Oualidia Lagoon Morocco	52.48	36.46	6.91	-	-	227.86	54.59	[35]
Black Sea	70–74	52–55	-	650–672	23–26	169–182	<0.1	[36]
Malaca Strait								
Dumai coast Indonesia	-	6.08	-	-	-	53.89	32.34	[37]
Bay of Bangal								
Pulicat lagoon S.E coast India	37	108	-	-	-	141	5	[38]
UCC	35	25		610	20	71	20	[39]
Average shale	90	45		850	50	95	20	[40]

Cu concentration shows wide variations ranges from 3 to 435 μg g^{-1}, with a mean value of 92 μg g^{-1} (Supplementary Table S1). The surface sediments (0–10 cm) show >250 μg g^{-1} Cu content (Figure 3g). The spikes are noted in the Cu profile at 21 and 30 cm depths, showing 340 and 535 μg g^{-1} Cu, respectively. A bulge in the trend line between 40 and 70 cm shows an increased Cu content in the sediments. Cu concentration steadily decreases down to the lowest (~3 μg g^{-1}) value in the bottom sediments (70–220 cm).

Ni concentration in the core sediments varies between 18.9 and 266.8 μg g^{-1}, with a mean value of 51.8 μg g^{-1} (Supplementary Table S1). Figure 3h displays a consistent Ni variation and shows > 50 μg g^{-1} Ni concentration in the upper half of the core and <50 μg g^{-1} in the lower section. In contrast to other metals, Ni content between 40 and 70 cm core depth does not show marked changes. The highest Ni (>250 μg g^{-1}) follows Cu peaks at ~20 cm and reflects an input over natural contribution. The only peak at ~110 cm with ~80 μg g^{-1} Ni is common to other investigated metals.

Pb varies between 16 and 332 μg g^{-1} with a mean value of 64 μg g^{-1} (Supplementary Table S1). The top 30 cm sediments of the core show ≥80 Pb μg g^{-1} (Figure 3i) with the highest Pb (332 μg g^{-1}) at ~30 cm. Pb content (≥200 μg g^{-1}) occurs between 40 and 60 cm depth. Like other metals, the Pb content is consistently low (<50 μg g^{-1}) in the lower section (110–220 cm) of the core.

3.4. Inter-Element Relationship

3.4.1. Matrix Correlation

Matrix correlation analysis was used to determine the genetic relationship between the elements and the controlling factors of metal distributions in the sediments (Table 2). The results showed that mud, LOI, and metals are positively correlated ($p < 0.05$), being very significant ($p < 0.01$), which suggests that organic-rich muddy sediments are the major control for metals distribution in the study area.

Table 2. Correlation matrix (core length = 220 cm, $n = 58$).

	Mud	Sand	Gravel	LOI	$CaCO_3$	Fe	Mn	Cr	Zn	Cu	Ni	Pb
			(%)						μg g^{-1}			
Mud	1											
Sand	−0.983 **	1										
Gravel	−0.792 **	0.665**	1									
Loi	0.314 *	−0.237	−0.487 **									
$CaCO_3$	−0.366 **	0.298*	0.494 **	−0.610 **	1							
Fe	0.417 **	−0.335*	−0.579 **	0.557 **	−0.850 **	1						
Mn	0.488 **	−0.414**	−0.604 **	0.582 **	−0.857 **	0.946 **	1					
Cr	0.177	−0.191	−0.085	0.422 **	0.05	0.126	0.197	1				
Zn	0.270 *	−0.218	−0.367 **	0.497 **	−0.557 **	0.707 **	0.673 **	0.320 *	1			
Cu	0.349 **	−0.274 *	−0.505 **	0.659 **	−0.732 **	0.810 **	0.0772 **	0.242	0.805 **	1		
Ni	0.119	−0.055	−0.298 *	0.249	−0.355 **	0.452 **	0.475 **	0.146	0.404 **	0.604 **	1	
Pb	0.225	−0.173	−0.337 **	0.630 **	−0.417 **	0.480 **	0.410 **	0.390 **	0.0753 **	0.805 **	0.238	1

**. Correlation is significant at the 0.01 level (2−tailed). *. Correlation is significant at the 0.05 level (2−tailed).

A significant inter-element correlation ($p < 0.01$) between Fe, Mn, Zn, Cu, Ni, and Pb suggests that Fe and Mn oxide/oxyhydroxides play an important role in metal accumulation Al-Salam Lagoon sediments. The Cr shows a significant correlation ($p < 0.05$, r = 0.32) with Zn and very significant correlation ($p < 0.01$, r = 0.39) with Pb. Cr is positively correlated with Fe and Mn. Core sediments from the Jeddah coast [29] also found a positive correlation between Fe-Mn and Cr. In this study, a positive correlation between Cr, mud, LOI, and metals implies a similar source of Cr input in the Al-Salam Lagoon. The negative correlation between $CaCO_3$, mud, and LOI with Fe, Mn, Ni, Pb, Zn, Cu, and Cr shows that these elements are not associated with $CaCO_3$. The $CaCO_3$ positively correlates with sand and gravel percentage, which signifies its association with gravel and sand. The inter-element correlations reflect that organic matter, sediment texture, and metals in the core Salam1 originated from the same source and were carried to the deposition site by muddy sediments.

3.4.2. Contamination Indices

The contaminant factor (CF) [41], geo-accumulation Index (I_{geo}) [42], and pollution load index (PLI) [43] were used to evaluate the heavy metal pollution in the core sediments. In this study, the sandy carbonate sediments represent natural background levels not affected by human activities. Hence, the background mean values of the metals present in the lower section (110–220 cm) of the Salam1 core (Table 1) were used to calculate the environmental indices. The results of the contaminant factor (CF), geo-accumulation Index (I_{geo}), and pollution load index (PLI) are tabulated in Table 3.

Table 3. Contamination factor (CF), geo-accumulation index (I_{geo}), and pollution load index (PLI) for the core sediments (Salam1) from the Al-Salam Lagoon, Jeddah coast. Average values for whole core (0–220 cm), polluted sediments (0–110 cm), and unpolluted sediments (110–220 cm).

Sediment Section		Fe		Mn		Cr		Zn		Cu		Ni		Pb		PLI
		CF	I_{geo}	CF	I_{geo}	CF	I_{geo}	CF	I_{geo}	CF	I_{geo}	CF	I_{geo}	CF	I_{geo}	
Whole core (0–220 cm)	Minimum	0.4	−2.1	0.5	−1.7	0.6	−1.4	0.2	−3.3	0.3	−2.6	0.5	−1.6	0.5	−1.7	0.5
	Maximum	6.6	2.1	4.5	1.6	1.9	0.3	5.9	2	36.3	4.6	7.2	2.3	9.5	2.7	6.7
	Average	2.1	0.2	1.9	0.1	1	−0.6	1.6	−0.3	7.6	1.1	1.4	−0.2	1.8	−0.1	2.4
Polluted sediments (0–110 cm)	Minimum	0.4	−2.1	0.5	−1.7	0.6	−1.4	0.2	−3.3	0.3	−2.6	0.5	−1.6	0.5	−1.7	0.5
	Maximum	6.6	2.1	4.5	1.6	1.9	0.3	5.9	2	36.3	4.6	7.2	2.3	9.5	2.7	6.7
	Average	2.9	0.7	2.5	0.5	1.1	−0.6	2.1	0	12.1	2	1.8	0	2.5	0.2	3.1
Unpolluted sediments (110–220 cm)	Minimum	0.4	−1.9	0.5	−1.6	0.7	−1.1	0.3	−2.4	0.6	−1.4	0.6	−1.4	0.6	−1.4	0.7
	Maximum	1.4	−0.1	1.9	0.3	1.1	−0.4	2.1	0.5	1.4	−0.1	1.2	−0.3	1.2	−0.3	1.8
	Average	1	−0.7	1	−0.6	1	−0.6	1	−0.8	1.1	−0.6	1	−0.6	1	−0.6	1.3

3.4.3. Contaminated Factor (CF)

The average contamination factor of the metals in the core Sa1 followed the order: Cu > Fe > Mn > Pb > Zn > Ni > Cr. The muddy sediments (0–110 cm) are highly contaminated with Cu (CF: 12) (Table 3). Pb, Zn, Ni, Mn, Cr, and Fe show moderate contamination $1 \leq CF < 3$, as proposed by Pecky [41]. Carbonate-rich sandy sediments (110–220 cm) show low (~1.0) CF. The CF profiles of metals exhibit a distinct upward increase from 70 cm depth (Figure 4). At about 30 cm depth, the increase in CF for Cu, Ni, Pb, and Zn ceases and becomes stable in the top surface sediments (Figure 4d,g). The Fe, Mn, and Cr CF show a continuous increase toward the surface. CF values (>1 to 20) between 70 and 30 cm depth show moderate to very high contamination. The CF, in the sediments above 40 cm, although, has decreased, CF (~20) for Cu still shows heavy contamination, and for other metals, CF ~2 shows moderate contamination. The pattern of Ni CF variations in the core sediments is subtle.

3.4.4. Geo-Accumulation Index (I_{geo})

The geo-accumulation index shows both negative and positive values (Table 3). The distribution of I_{geo} values along the core (Figure 5) exhibits a progressive upward increasing trend from 100 cm depth. I_{geo} negative values steadily increase upward to ≥1 for Fe, Mn, Zn, and Ni until the 70 cm depth. For Cr and Pb, the I_{geo} values remain low (−1 to −0.5) at this depth. A significant upward increase in I_{geo} is observed between 70 and 50 cm depth. In the top 40 cm sediments, the I_{geo} values for Fe, Mn, Zn, and Pb are 1–2, whereas Ni and Cr show values between −9 to 0 (Figure 5c,e). Cu shows the highest (~4.0) I_{geo} values in the top 40 cm sediments (Figure 5f).

3.4.5. Pollution Load Index (PLI)

The average pollution load index value for the Salam1 core sediments is 2.4 (Table 3). The PLI values for the muddy sediments (0–100 cm) and carbonate-rich sediments (110–220 cm) of the core are 3.1 and 1.3, respectively. PLI value 3 for the muddy sediments is similar to the reported PLI (3.29) from the Jeddah coast [29]. Figure 6 shows that the PLI value increases from 1.0 at ~100 cm to 5 at 50 cm sediment depth. The pollution load index increased exponentially between 70 and 50 cm depth. From 50 cm to the surface, the PLI value varies between 3 and 7, indicating heavy pollution. According to the pollution grades of Tomlinson et al. [43], these sediments are classed as extremely heavily polluted. The bottom sediments (70–220 cm) in the core display uniform low (~1) PLI values.

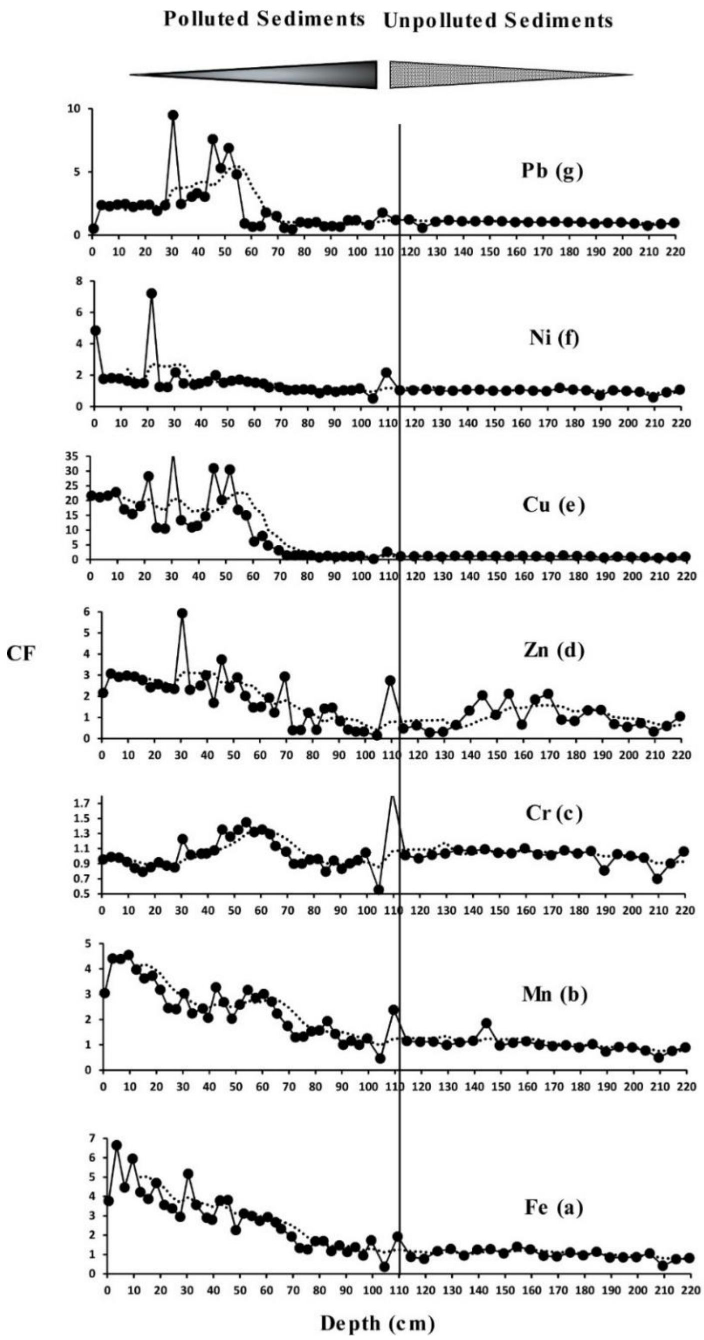

Figure 4. (**a**,**b**) Vertical distribution of contamination factor (**c**–**g**) for metals in the Salam1 core from Al−Salam Lagoon along the Jeddah coast, Saudi Arabia. (Trend line of 5 Average is shown by dots).

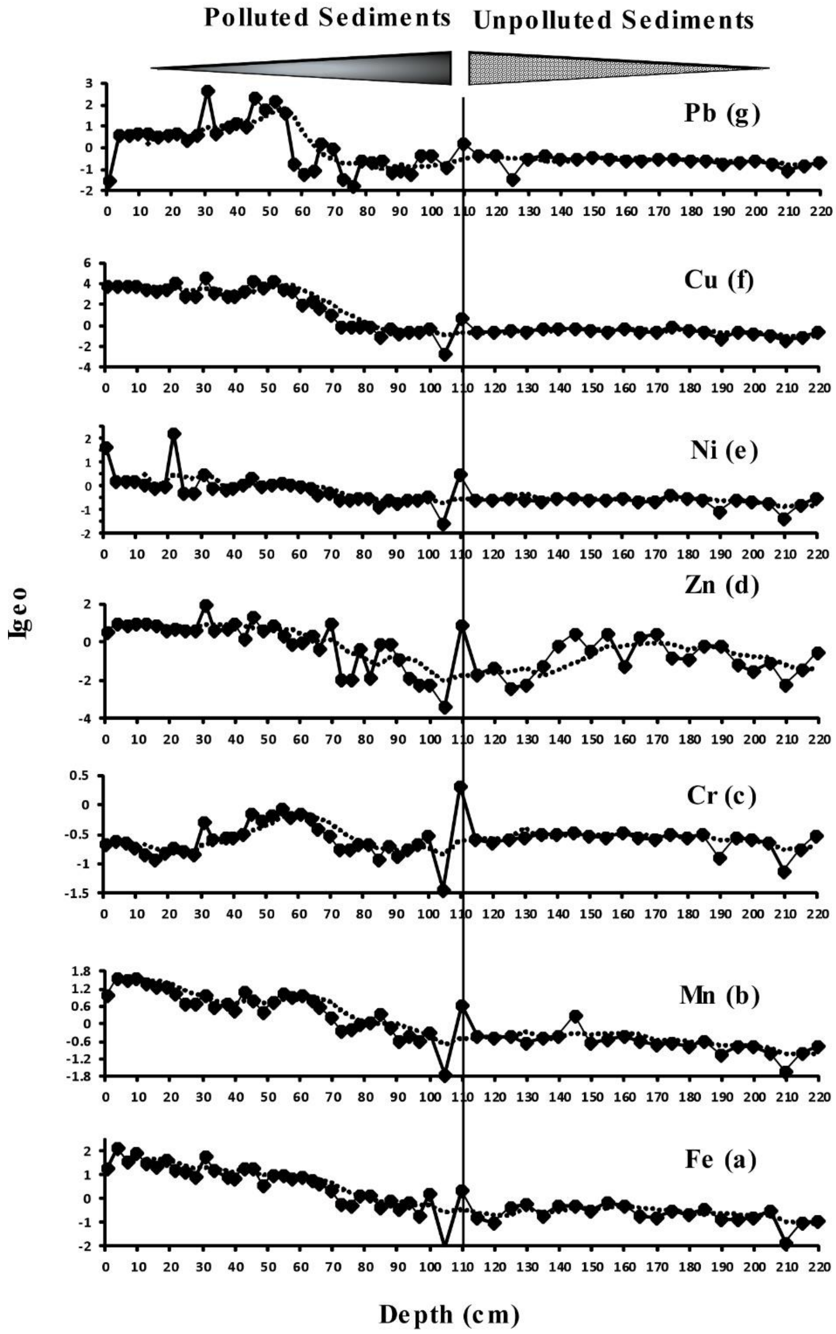

Figure 5. (**a**–**g**) Profiles of geo-accumulation index (I_{geo}) for metals in the Salam1 core from Al-Salam Lagoon, along the Jeddah coast, Saudi Arabia. (Trend line of 5 Average is shown by dots).

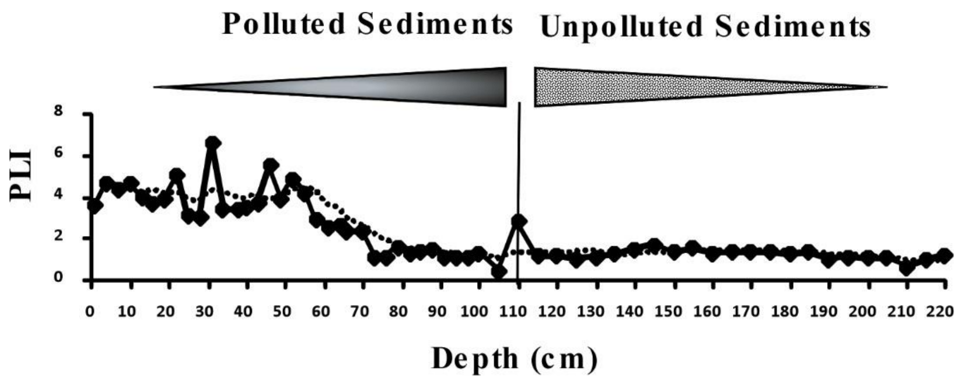

Figure 6. Pollution load index (PLI) variations in the Salam1 core from Al-Salam Lagoon along the Jeddah coast, Saudi Arabia. (Trend line of 5 Average is shown by dots).

4. Discussion

The Al-Salam lagoon is a recreational site in the central part of the Jeddah coast. However, four outfall pipes of sewage and industrial wastes discharge in it. Therefore, it has received a large amount of domestic/industrial wastewater with around 100,000 m^3 daily [20]. These waste materials are generally enriched by heavy metals that may cause critical pollution in the lagoon. The results showed two distinct sections throughout the core, classified as polluted and unpolluted sediments in the Al-Salam core. The sediment types in the core confirm the previous studies [14,44], which showed that Jeddah coastal sediments are largely carbonate sand and polluted muddy sediments show human impacts. The contaminated sediments are the black sludge mud at the top of the core deposited during the industrial time that probably started in 1970.

In contrast, the carbonate-rich sediments (110–220 cm) at the down section of the core were deposited in a pre-industrial time, probably pre-1970 [22]. The variations in sediment texture along the core reflect an evolutionary trend of the anthropogenic impacts as inferred from the associated organic matter and metals concentration and geochemical indices. In this study, a significant positive correlation between mud, organic matter, and metal content reflects an intimate relationship, suggesting texture control on both organic matter and metal enrichment [28]. El-Sayed and Basaham [28] reported higher metal content in the muddy sediments from the Jeddah coast. An increase in mud from 70% to 90%, together with organic matter from 5% to 20%, in the upper 70 cm sediments of Salam1 core show a high flux of mud and organic matter.

Metal concentration in the muddy sediments (0–110 cm) is higher than the published data from the Gulf of Aqaba [11], Persian Gulf [34], Mediterranean Sea [9], Malaca strait [37], and Bay of Bengal [37] (Table 1). Cr content in the Black Sea sediments [37] is slightly higher than in the Al-Salam Lagoon sediments. When compared with the surface sediments of the Red Sea coast [3,30,32], except Fe and Mn, metal concentration is high (Table 1). Along the Jeddah coast, Al-Shabab Lagoon and Sharm Obhor sediments show high Cr [14,31]. Al-Mur et al. [29] reported a high concentration for Fe, Cr, Mn, Cu, Zn, Ni, and Pb from a location close to the study area (Table 1). This shows a substantial metal input along the Jeddah coast, attributed to the human activities resulting from urbanization. Bellucci et al. [45] and Bertolotto et al. [46] reported high metal concentration in the surface sediments, showing the impacts of human activities. Badar et al. [16] and Al-Mur et al. [29] linked the concentration of the elevated metals in sediments from Jeddah and other coastal areas of the Red sea with anthropogenic activities. Abu-Zied et al. [14] reported high metal concentration in sediments from Al-Arbaeen and Al-Shabab lagoons along the Jeddah coast.

The positive inter-element correlation between metals suggests that they are sourced from a common anthropogenic input. After being released from the source, it is carried by the fine-grained sediments to the depositional site. Urban waste from the Jeddah coast is loaded with metals derived from sources such as power plants, municipal discharge, desalination plants, refinery plants, and harbor activities. Fe, Cr, and Mn sources are linked to iron and steel industries and ship wreckage. The steel contains Cr and Mn, which are added as an anticorrosive material [47]. Ni is used as a catalyzing agent in the oil refinery processes. Cu was introduced through antifouling paints from a ship. Zn in sediments is mostly associated with sewage, industries, food wastes, and antifouling paints [48]. Kadi [49] found 450 $\mu g\ g^{-1}$ Zn in the road dust of the Jeddah urban areas. In coastal waters, Cu present in the urban run-off is linked with sewage sludge, dumpsites, municipal wastes, and antifouling paints [50]. Fungicides and wood preservatives also contribute a high level of Cu, especially for public health [51].

The similarity of the Cu and organic matter (LOI) variation trends along the Salam1 core sediments (Figure 3b,g) suggests an anthropogenic source associated with sewage discharge loaded with high organic matter. The Cu spikes in the top 40 cm sediments could result from flash floods in the study area, which washed enormous urban waste material from the inland and transported along the coast. Pb in the core sediments follows other metals and indicates sources such as fossil fuel combustion, ship industries, and road traffic. In the marine environment, Laxen [52] and Abu-Hilal [53] linked Pb to boat exhaust, oil spillage, fishing boats, and sewage effluents. Usman et al. [54] indicated a high Pb level in the Red Sea from the use of fossil fuel. Banat et al. [55] found Pb contamination from automobile exhaust to the highway areas. The soils of the Jeddah roadsides contain 105 $\mu g\ g^{-1}$ Pb [49]. Studies [14,16,31,56] from the Jeddah coast suggested Pb contamination from boat exhaust, leakage of oil from boats, and sewage effluents. Likewise, elevated ($\geq$250 $\mu g\ g^{-1}$) Pb content in the sediments section (30–60 cm) of the Salam1 core shows contamination from similar sources as suggested in earlier studies.

The fluctuations of Fe and Mn along the Salam1 core suggest a characteristic geochemical behavior of Fe and Mn. In a reduced environment, precipitation of overlying Fe-rich water or upward movement of pore water results in high Fe in the sediments [57]. Fe oxy-hydroxides dissolve in a reducing environment [58], move upward, and precipitate in sediments' oxic–suboxic zone. The Mn distribution in the core sediments is in conformity with the results of Pattan [59] and Badr et al. [16]. Mn subsurface maxima and downward decreasing Mn in the sediments reflect that pore water from reducing zone supply Mn^{2+} and oxidized to Mn^{4+} at the surface [59]. Following Janaki-Raman et al. [60], a higher Mn concentration in the upper 30 cm sediments of the Salam1 core shows an upward movement of Mn. A decline in Mn at the surface suggests that Mn is lost to the overlying water at the sediments–water interface, as showed by high Mn in near-surface waters along the Jeddah coast [61]. Cr in the upper 30 cm sediments is ~50 $\mu g\ g^{-1}$. Badr et al. [16] obtained a similar Cr profile from Jeddah and other coastal areas of Saudi Arabia. Cr is a redox-sensitive element, and Pattan et al. [62] suggested co-precipitation of Cr with Mn-oxyhydroxide. Following Janaki-Raman et al. [60], the Cr distribution in the core Salam1 appears to show a diagenetic control. Gaillard et al. [63] showed that Cr as (Cr^{6+}) being a mobile element accumulates in the reduced sediments.

In polluted sediments, higher Zn content is largely associated with a reducible fraction [64–67]. Studies showed Zn bonding with oxy-hydroxides [68] and Fe and Mn oxides as a carrier of Zn [64,69]. The strong positive correlation ($r \geq 0.67$) of Zn with Fe and Mn in the investigated sediments of the Salam1 core supports this (Table 2).

The Cu concentration variations show a close similarity with LOI and other metals. The strong positive correlation of Cu with LOI (Table 2) shows its affinity for organic matter [41]. Studies showed high concentrations of Cu in organic-rich sediments [42,70–72]. A high ($\geq$300 $\mu g\ g^{-1}$) Cu concentration in the upper section (100 cm) of the Salam1 core sediments is like the published results from the Jeddah area [16,29].

Ni is a common crustal element and, through various processes, includes rock weathering, erosion, atmospheric fallout, and industrial wastes introduced into the environment [73]. Badr et al. [16] reported high Ni sediments from an area close to the oil refinery and suggested a linkage with the oil refinery processes. The high Ni > 250 $\mu g\ g^{-1}$ at 20 cm depth in Salam 1 core also reflects an input in excess of natural contribution.

The metal enrichment variations in the sediments along the Salam1 core reflect an evolutionary trend of the anthropogenic impacts. Along the Jeddah coast, urbanization and accompanying developmental activities increased during the last few decades. This increased coastal pollution. The elevated concentration of metals in the muddy sediments present in the upper half of the Salam1 core suggests that Fe, Mn, Cr, Ni, Zn, Cu, and Pb were introduced into the Al-Salam Lagoon from the land-based sources, washed through as urban waste, and deposited in the coastal waters.

The carbonate-rich sediments (110–220 cm) show an unpolluted environment. From ~100 cm depth, a steadily upward increase in metal concentration shows the quantities of effluents loaded with contaminants discharged and transported over time. This reflects the introduction of metal pollution to the Al-Salam Lagoon, which increased with urbanization. Because of increased industrial and human activities, metal concentration was found to be higher in the surface layers than in the deeper sediments [45,46].

Contamination Evaluation

The sandy carbonate sediments (110 and 220 cm) in the core show a negative geo-accumulation index (I_{geo}) and low (~1.0) CF. This implies that the deeper sediments (110–220 cm) are unpolluted. The I_{geo} value 2 for Cu in the organic-rich muddy sediments (0 to 100 cm) indicates moderate contamination and falls in class 2. The I_{geo} value (0–1) for Fe, Mn, Zn, Ni, and Pb indicates unpolluted to moderately polluted sediments. Pb profile (Figure 5g) shows high ($\leq$3) I_{geo} values in sediments between (40–60 cm), which suggests moderate-to-heavy Pb contamination.

Consistent trends of metal concentration, CF, and I_{geo} variations along the core suggest a common pollutant source and the pollution load variations over time. The PLI value and consistent trend show a progressive evolutionary trend from a depth of 70 cm. PLI value increased exponentially from 2 and reached $\geq$5 at 50 cm depth, followed by a decline to 4 to the surface. In the muddy sediments above 70 cm, the PLI value ranges between 3 and 7, indicating heavy pollution. The stable trends of the environmental indicators (CF, Igeo, and PLI) in the top 40 cm sediments probably indicate compliance with environmental regulation to reduce the discharge of effluents in the Jeddah coastal area. Honsono et al. [74] reported similar reduction trends in metal pollution elsewhere due to environmental legislation. The sewage had polluted Jeddah's coastal waters since the urbanization time. The sewage dumping was stopped in the 1990s [20]. Risk et al.'s [22] $\delta^{15}N$ study of the black corals of Jeddah showed increasing sewage discharge levels since the 1950s, which decreased in the 1980s. A stable, consistent low trend of Cu, Zn, Pb, CF, I_{geo}, and PLI in the top ~40 cm sediments support the findings of Risk et al. [22].

A low concentration of the metals in the deeper carbonate-rich sediments between 110 and 220 cm suggests a pristine environment not contaminated by land-based activities. Besides the environmental indices (CF, I_{geo}, and PLI), a positive correlation between sand and $CaCO_3$ content and a negative correlation of sand with organic carbon (LOI) and metals content in the Salam1 core sediments support this. The negative geo-accumulation index (I_{geo}) in the carbonate-rich sediments implies that metals are geogenically sourced, and a low (~1) uniform PLI value shows pollution-free sediments.

5. Conclusions

The texture, organic carbon (LOI), metal concentration, and geochemical indices variations in the sediments along the Salam1 core reflect an evolutionary trend of pollution along the Jeddah coast. The positive, statistically significant ($p < 0.05$) correlation between mud, LOI, and metals indicates that they are genetically associated and are influenced by the same factors and processes. A significant positive correlation ($p < 0.05$, r = 0.31) between mud and LOI suggests a close association of grain size with organic matter. In the upper half of the core, the organic-rich mud with high metals shows increased metal input. A positive inter-element correlation and a similarity in the metal distribution down the core suggest a common source. Contaminants washed as surface run-off from inland

and discharged into the coastal area. The degree of contamination (CF) suggests high Cu contamination with (CF: 12) and moderate Pb, Zn, Ni, Mn, Cr, and Fe contamination. The PLI value (~3) grades the sediment as heavily polluted. In the lower half (110 cm 220 cm) of the core, the carbonate-rich sandy sediments are low in metal content and LOI%. The negative geo-accumulation index (I_{geo}) implies that metals are geogenically sourced, and low (~1) CF and PLI values suggest no impacts of anthropogenic activities.

Consistent trends of metal concentration, CF, and I_{geo} variations along the core suggest a common pollutant source with varied pollution load over time. Progressive developmental activities in the study area and subsequent environmental impacts in the Al-Salam Lagoon core sediments are reflected by the sediment pollution as the mud increases, followed by metal pollution. The variations in environmental indices (CF, I_{geo}, and PLI) indicate varying pollution load or inland usage of contaminant source materials along the Jeddah Coast.

A distinct upward increase in the organic carbon (LOI), metal concentration, CF, I_{geo}, and PLI in the sediment section from 70 to 40 cm suggests uncontrolled pollution. The metal enrichment and contamination level decline from 40 cm above and become stable in the surface sediments. In the core sediments above 40 cm, the invariable trend for Cu, Zn, Pb, CF, I_{geo}, and PLI suggests that metal pollution along the Jeddah coast is presently controlled after an exponential increase in the past decades. Decreasing and the stable trend in the top 40 cm sediments is consistent with the measures taken in the recent past to stop the direct discharge of municipal sewage loaded with urban waste into the Jeddah coastal zone.

Supplementary Materials: The following are available online at https://www.mdpi.com/article/10.3390/jmse9080899/s1, Table S1: Mud%, Sand%, Gravel%, LOI%, $CaCo_3$%, and heavy metals concentration (Fe, Mn, Cr, Zn, Cu, Ni, and Pb) in the core SALAM1 sediments from Al-Salam Lagoon, Jeddah coast-Saudi Arabia.

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