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Soil & Sediment Contamination, 13:299–311, 2004
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ISSN: 1058-8337 print
DOI: 10.1080/10588330490445321

An Overview of Metal Pollution in the Western Harbour of Alexandria, Egypt

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Limited data are available on the concentration of metals in sediments in the Western Harbour of Alexandria. The most comprehensive record is from a survey conducted more than a decade ago. Industrial and human activities in and around this area have increased dramatically in the last 20 years. The purpose of this study was to determine the concentrations of heavy metals in surfacial bottom sediments of the harbor, to assess their potential biological effects and to identify their possible sources. Sediment samples from 21 stations throughout the harbor were analyzed for grain size, total organic carbon content (TOC), and metals (Al, As, Ba, Be, Cd, Cr, Cu, Fe, Mg, Mn, Ni, Pb, Se, Sn, V, and Zn) to assess the extent of contamination in the area. The results indicated that concentrations of metals in the sediments varied widely depending on the location. High levels of metals were observed in the Arsenal Basin and the outfall area of El Mahmoudiya Canal in the inner harbor. The concentrations of metals were found to be higher than those recorded in the previous study. However, with some exceptions, most of the changes in the metal concentrations could be accounted for by the variations in aluminum, which represents the variations in mineralogy and grain size, indicating that the majority of the metals were of "natural" origin. The present data were also compared with results from other areas.

Keywords Metals, harbor sediments, pollution, sources, Alexandria, Egypt.

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Introduction

The Western Harbour of Alexandria, situated to the west of the city of Alexandria along the Mediterranean coast of Egypt (Figure 1), is a complex harbor with an area of about 31 km^2. The harbor is connected to the open sea by a narrow strait, which protects the bay from the prevailing NW winds and renders the harbor a safe anchorage. The water body inside the harbor has an average depth of 7 m. The harbor was naturally formed during the Pre-Holocene subsidence of the coast and the subsequent transgression of the sea (Butzer, 1960). Calcareous marine deposits cover the continental shelf surrounding the harbor.

The Western Harbour of Alexandria is the major trade port of the Northern Territory of Egypt. Industrial activities have increased dramatically in this area over the past 20 years. The harbor handles approximately 75% of all ship-borne cargo of the country. As a result of aggressive urbanization and industrialization of the Alexandria region, the coastal waters in general and the Western Harbour in particular have received considerable amounts of treated and untreated industrial, agricultural and domestic wastes (Salem and Sharkawi, 1981). These wastes are derived principally from effluent discharged from the El Mex pumping station to the west of the harbor. The effluent consists of overflow from Lake Maryut

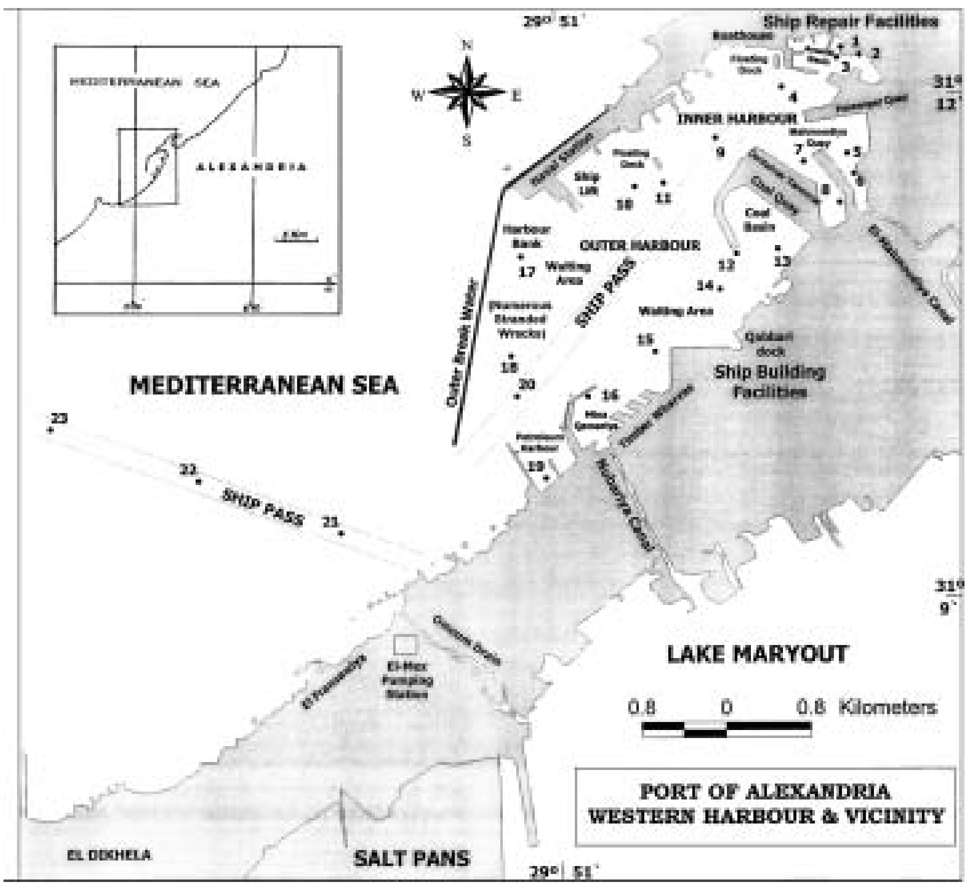

Figure 1. Location map of study area.

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(a coastal lagoon heavily polluted mostly by domestic and industrial wastes), and the drainage water from the El-Umum drain and El-Noubariya Canal. Industries that benefit from their quay-side location include a chlor-alkali plant (Misr Chemical Industries). Portland cement factories discharge unknown quantity of wastes into the harbor. Minor amounts of industrial wastes are directly discharged into the harbor through tanneries.

Because the harbor is a semi-closed basin with restricted water circulation, it may serve as an entrapment of the wastes introduced from land-based sources as well as from the harbor itself due to shipping activities. Future development and continued operation of Alexandria Harbour is of great economic importance to this region, but these activities may also impact its ecological functioning.

Little data are available on the levels of metals in the western harbor. The most comprehensive records were from a survey done about 13 years ago (El-Sayed *et al.*, 1988). It was therefore deemed necessary to set up a monitoring program to determine the current concentrations of metals in bottom sediments, and to identify any area where high concentrations of potential hazardous contaminants were present in the harbor.

Materials and Methods

Sediment samples were analyzed for trace metals at the Geochemical and Environmental Research Group, Texas A&M University. Matrix spikes, laboratory sample duplicates, and laboratory blanks were processed with each batch of samples (10 samples/batch) as quality control samples. Duplicates were produced by sub sampling in the laboratory. Standard reference materials (National Institute of Standards and Technology) were analyzed to audit the performance of the analytical methods. The quality assurance standards are those of the NOAA's National Status and Trend Program, US EPA's Environmental Monitoring and Assessment Program-Near Coastal (EMAP-NC) and the U.S. Fish and Wildlife Service (FWS) for trace contaminant analyses (Wade *et al*., 1988). These methods have undergone extensive intercaliberation with EPA, NOAA, NIST and FWS. Detailed methods are provided elsewhere (Kennicutt II *et al*., 1992).

Samples Collection. Sediment samples were collected in April 1999 at 21 locations (Figure 1). Station locations were chosen to provide good areal coverage. Samples were taken with an Ekman grab sampler. All samples were carefully inspected to ensure that undisturbed sediments were collected.

Trace Metals. The major analytical technique used for trace metal determination was atomic absorption spectrophotometry (AAS) in the flame mode and inductive coupled plasma spectrophotometry (ICP) for those elements in high concentrations. Graphite furnace (GF/AAS) was used for the determination of both selenium and arsenic. Samples were pressure-digested in 50-mL closed all-Teflon "bombs" (Savillex Co.; Brooks *et al*., 1988). Sediment aliquots (ca. 200 mg) were digested at 130°C in a matrix of nitric, perchloric, and hydrofluoric acids. A saturated boric acid solution was then added to complete the dissolution.

Organic Carbon and Grain Size. Organic carbon was measured following flash combustion in an enriched atmosphere of oxygen using a LECO carbon analyzer. Samples were acidified using dilute HCl in methanol and then dried. Particle size analysis was conducted by laser granulometry (Mastersizer/E, Malvern Instruments), and the results summarized using the graphical methods of Folk (1974).

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Results and Discussion

Spatial Distribution of Metals

The combined effect of relatively deep water and weak tidal currents indicated by moderately sorted sediments in the inner harbor allows the deposition of greater than 90% of fine grained sediments that were smaller than 63 μm in particle size. This area was surrounded by bands of progressively coarser sediments with poorly sorted sandy sediment at the outlet of the harbor.

Because the surface area of sediments is grain-size dependent and controls the adsorption of metals in the sediment (Mayer and Fink, 1980), the concentrations of some trace metals correlate with the clay fraction of the sediment that provides the greatest surface area for the adsorption of metals (Jickells and Knap, 1984). However, interpretation of metal concentrations in sediments is compounded by the metal's natural concentration in different minerals and by the relationship of metals to sediment grain-size characteristics.

Trefry and Presley (1976), Windom *et al.* (1989), and Summers *et al.* (1996) presented a simple method of distinguishing natural changes from those induced by anthropogenic activity. Metal concentrations were normalized to aluminum or iron to determine whether a sediment sample was enriched with metals when compared to the sample's "natural" conditions. Because Al is tightly associated with aluminosilicate fraction that is the dominant metal-bearing phase of the sediment, i.e., clay/silt fraction of the sediment (e.g., Figure 2), Al was used as a normalization factor in predicting the enrichment or depletion of heavy metals in sediment. Contaminated samples have been identified by plotting metal concentrations against Al concentrations (Windom *et al.*, 1989). On a scatter plot, the data points that fall within 95% confidence levels of the data base were be taken to be natural, and those points that were above the confidence limit were considered to be enriched (Windom *et al.*, 1989).

There was a significant correlation between the aluminum concentration and % silt/clay, and between the aluminium concentration and % $<$63 μm fraction (Figure 2). However, no statistically significant correlation was observed between organic carbon and trace metals.

Metal distributions correlated with the distribution of fine-grained clays. The highest concentrations for most of the metals were found in the inner harbor, and metal

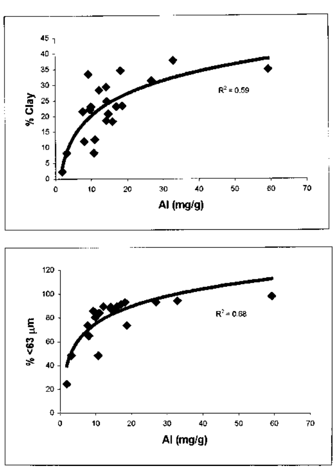

Figure 2. Relationship between particle size distribution and clay content vs aluminum concentration in the sediment from the Western Harbour of Alexandria.

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concentrations progressively decreased towards the outlet of the harbor. There was a similar distribution pattern of fine-grained sediment. Most of the metals studied showed the highest metal/Al ratios near the inlet of the Arsenal Basin (stations 1, 2, and 3) and the Boathouse (station 4), near the outfall of the El Mahmoudiya Canal (stations 5 and 6), as well as the inlet between the El Mahmoudiya Quay and the Container Terminal (stations 7 and 8; Figure 3).

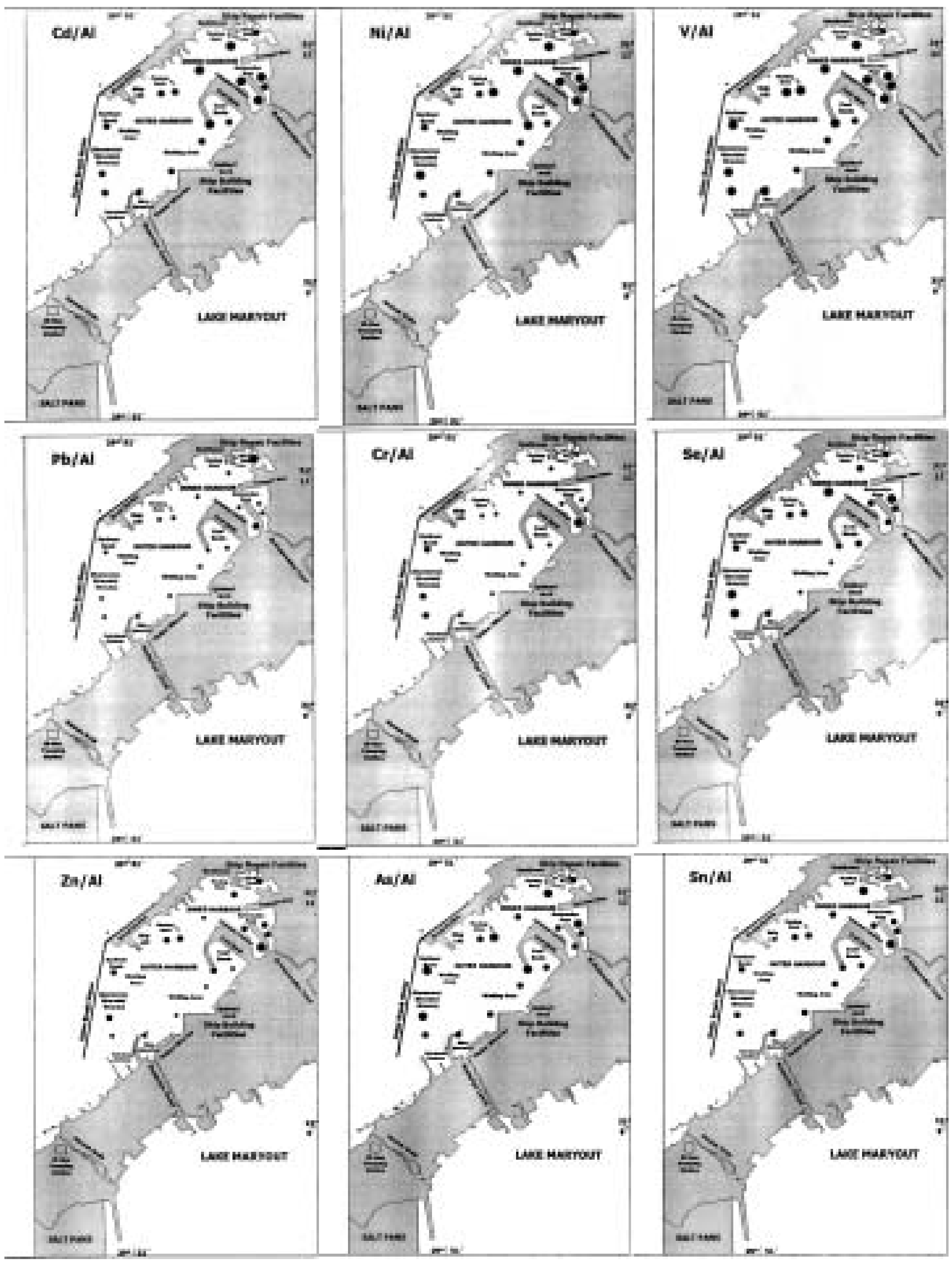

Figure 3. The distribution of metal/Al ratios in the Western Harbour of Alexandria; the size of the circle denotes the value of the ratio (● >0.75, ● 0.25–0.75 and • <0.25).

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All these stations are located near wastewater effluent areas in the inner harbor. The spatial distribution of high metal Al ratios closely correlate with the distribution of fine clay in the sediment, which were associated to the areas of outfalls and discharges of agricultural runoff and industrial effluents (Figure 3).

Cadmium and lead are highly toxic metals widely used in diverse industrial processes. Concentrations of Cd in most of the samples from the inner harbor were high. Concentrations of Cd in the sediments near the outlet of the harbor are relatively low (stations 19 and 21 with 0.45 and 0.83 μg g^{-1}, respectively, Table 1). However, the correlation between Cd and Al is significant (r^2 of 0.61, Figure 4), suggesting that most of the Cd detected in the samples was associated with aluminum enriched clay minerals (Summers *et al.*, 1996) and co-varies with Al (Figure 4).

The presence of high concentrations of Cd in the sediments of the Western Harbour appears to be directly related to urban and industrial runoff discharged from the El

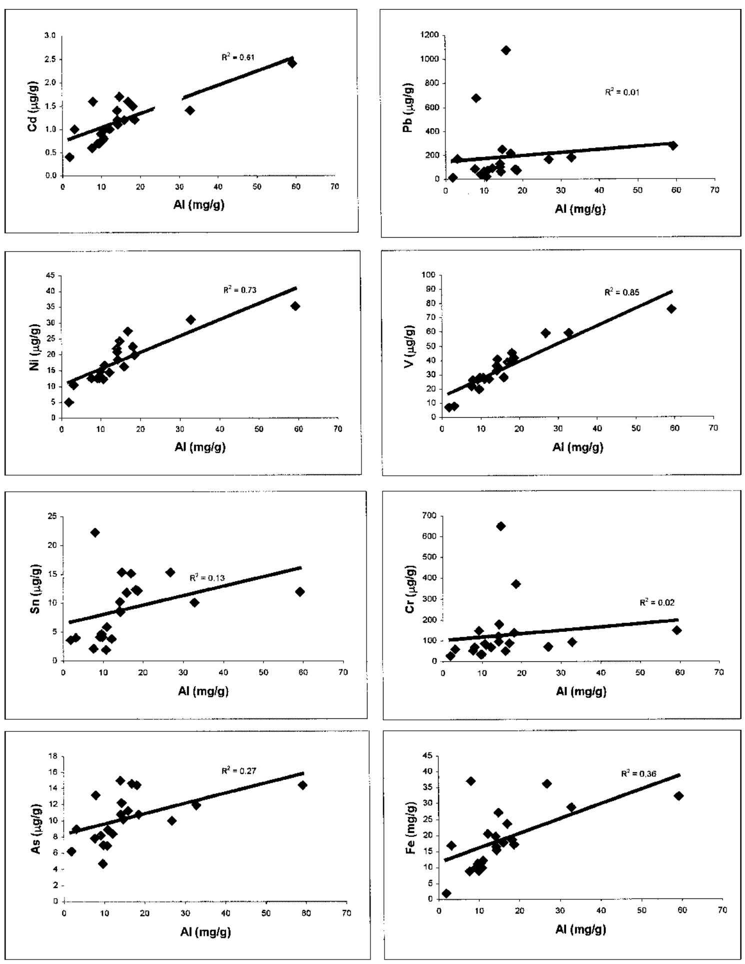

Figure 4. Relationship of trace metal concentrations (μg/g, dry weight) with that of Al (mg/g, dry weight) in the sediment from the Western Harbour of Alexandria. Scattering in data points, particularly in the low Al concentration range, indicates extraneous input of trace metals to the sediments.

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Table 1
Concentrations of metals ($\mu g\ g^{-1}$ dry weight) in sediments of Alexandria Western Harbour

Station	Location	% TOC	% <63 μm	phi	Al	As	Ba	Be	Cd	Cu	Cr	Fe	Mg	Mn	Ni	Pb	Se	Sn	V	Zn
1	Arsenal basin	11.3	64.6	3.0	8011	13.1	728	0.7	1.6	378	68	36953	14416	232	39.7	678	2.8	22.2	26.1	652
2	Careening basin	3.6	48.4	3.2	3145	9.0	321	0.3	1.0	37	59	16853	16817	176	10.5	170	2.8	4.0	7.7	1221
3	Arsenal basin	7.9	89.3	2.3	15915	11.2	294	1.2	1.2	136	49	17696	14067	216	16.3	1070	2.9	11.8	27.9	282
4	Boathouse	6.8	91.3	1.8	16940	14.6	152	0.9	1.6	134	87	23568	13983	204	27.3	215	3.0	15.1	38.7	382
5	El-Mahmudiya quay	5.6	83.9	2.7	10937	8.9	127	0.5	1.0	66	79	12159	14327	147	16.6	71	3.4	5.9	27.0	211
6	El-Mahmudiya quay	3.1	93.1	1.6	26795	10.0	1155	1.3	1.8	168	70	36140	16706	278	52.8	165	2.9	15.3	59.1	349
7	Container terminal	3.5	87.8	2.2	14154	10.8	260	0.7	1.2	102	95	16462	13896	176	20.8	101	3.2	10.2	32.8	268
8	El-Mahmudiya quay	5.3	87.5	2.5	14735	10.2	2593	0.8	1.7	125	649	27104	13871	255	24.3	250	3.2	15.3	34.3	507
9	Ship pass (inner harbour)	8.6	80.2	2.9	9873	7.0	77	0.5	0.9	27	33	8878	13036	109	14.7	63	3.0	4.1	27.7	58
10	Ship lift	8.3	92.8	1.9	18149	14.4	119	0.8	1.5	84	138	18572	16235	202	22.5	83	3.2	12.3	45.1	194
11	Ship pass (outer harbour)	3.8	73.6	3.0	7679	7.8	73	0.4	0.6	30	51	8819	14279	143	12.5	85	1.4	2.1	22.0	70
12	Coal quay	7.8	88.9	2.2	14097	15.0	400	0.8	1.4	79	120	19684	14919	199	21.9	128	3.1	8.7	36.4	203
13	Coal basin	23.4	84.8	2.6	9635	4.7	175	0.6	0.7	28	33	11134	6591	139	12.5	43	1.7	4.7	19.7	65
14	Mooring area (1)	13.4	98.1	1.4	59204	14.4	370	2.6	2.4	207	146	32150	11984	252	35.2	276	2.8	11.9	75.6	269
15	Mooring Area (2)	10.3	94.1	1.7	32760	11.9	198	1.7	1.4	58	92	28655	16602	317	30.9	180	0.8	10.1	59.1	190
16	Mina El-qamariya	18.5	89.4	2.0	12144	8.4	195	0.6	1.0	36	67	20506	16227	186	14.4	91	3.2	3.8	26.8	88
17	Harbour bank	7.4	88.7	2.2	14328	12.2	102	0.7	1.1	70	178	15296	15941	166	18.4	63	2.9	8.5	40.7	180
18	Stranded wrecks	6.4	85.7	2.1	9191	8.2	87	0.4	0.7	39	147	9977	15665	159	12.5	38	3.3	4.1	26.2	105
19	Petroleum Harbour	5.7	24.4	3.6	1867	6.2	59	0.1	0.4	30	26	1865	1277	91	5.0	33	2.1	3.6	7.0	33
20	Harbour inlet (inside)	3.4	73.5	3.1	18620	10.8	84	0.9	1.2	42	371	17030	16025	275	19.9	88	1.2	12.1	41.7	88
21	Harbour inlet (outside)	2.3	48.2	3.4	10684	6.9	64	0.5	0.8	29	84	9879	13350	209	12.3	21	2.3	1.9	27.8	48

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Mahmoudiya Canal (Figure 3). The sediments near the runoff areas were mostly fine-grained clayey mud with <63 μm fraction consisting of $>90\%$ of the sediment. Both Al and Cd concentrations were high in these sediments deposited near the runoff and discharge area.

Total Ni and V concentrations in the sediments of the Western Harbour follow a relatively uniform pattern, as their values range from 5.02 to 39.66 μg g^{-1} for Ni and from 7 to 75.6 μg g^{-1} for V (Table 1). Excluding two samples from Arsenal Basin and El-Mahmoudiya Quay that had high concentrations of Ni and V, both Ni and V had strong positive correlation with Al in all the samples (r^2 of 0.73 and 0.85 for Ni and V, respectively; Figure 4).

The outer harbor waiting area (stations 14 and 15), stranded wrecks area (station 17), the ships waiting area near the Ship Lift (station 10), the area to the outfall of the El Mahmoudiya Canal and the sewage outfall in the Customs Administration Quay (Station 6), have elevated concentrations of Ni and V (Table 1 and Figure 3). Since Ni and V are components of crude oil and Ni is used as a catalyzing agent in the oil refining process, these elevated concentrations of Ni and V may be related to petrochemical industries such as Alexandria Petroleum Company at El Mex that have been active for several decades, and their wastes in the past were discharged directly to the ocean without any prior treatment.

Correlation between Al and Pb, Sn, Cr, As are poor, with correlation coefficient r^2 ranging from 0.20 to 0.43 (Figure 4). Most of the scattering in the metal concentrations occurs in the low Al concentration range. The large scattering in metal concentrations (particularly the high metal concentrations relatively to Al) suggests a possible input of anthropogenic metals to these samples.

Sediment samples from locations in the narrow inlets of the inner harbor (Stations 1 and 3) of Arsenal Basin had the highest Pb concentrations (677.8 and 1070 μg g^{-1} for stations 1 and 3, respectively). The high Pb concentration may be related to the continuous and sometimes massive runoff of residential wastewater into the sea, as well as the atmospheric emissions (leaded gasoline) from urban and industrial areas of west Alexandria city. Williams (1995) suggested that anthropogenic lead is mostly associated with the slowest settling fraction of particulate matter, and so has a potential to be transported far from its source and not settle to the sediment until reaching areas of high water stability. It is possible that anthropogenically derived lead is being transported to these near shore areas and deposited to the bottom sediments in the regions of weak currents.

Arsenic concentrations in sediments ranged from 4.72 to 14.97 μg g^{-1}. Elevated concentrations in the sediments can be seen near the Ship Lift (station 10), and the quay for woods (stations 12 and 14, Figure 3). The narrow inlets of the inner harbor also have elevated concentrations of arsenic (Stations 1 and 4). A weak positive correlation between As and Al indicates that natural As was an important component in addition to the anthropogenic As (Figure 4).

Two samples had high chromium concentrations. Samples from station 8 (649 μg g^{-1}) in the inner harbor and from station 20 (371 μg g^{-1}, near the outlet of the harbor) had high concentration of chromium compared to the other stations (Table 1). Correlation between Cr (even with the excluding of the two samples from station 8 and station 20 that had high Cr concentrations) and Al is weak (Figure 4), indicating that anthropogenic input of Cr was significant.

The concentration of selenium was found to be in the range of 0.79–3.45 μg g^{-1}. Most samples from the outer and the inner harbor have Se concentrations more than 2 μg g^{-1}. The high levels of Se may be attributed to the anthropogenic inputs from leaded gasoline,

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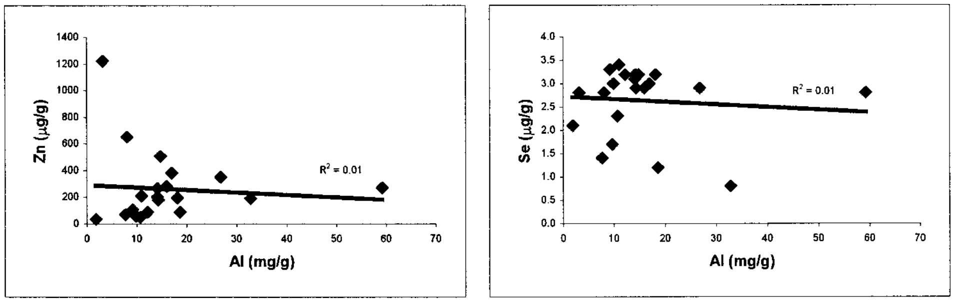

Figure 5. Poor negative correlation between Al and Se, Zn in the sediment from the Western Harbour of Alexandria indicates anthropogenic input of Se and Zn to the sediments.

ship exhaust systems and oil refinery waste streams (Garbisu *et al.*, 1995). Correlation between Se and Al is poor and most of the samples with high Se concentrations had low Al concentration (Figure 5). Similarly, most of the samples with high Zn concentrations had low Al concentrations (Figure 5). The poor negative correlation between Se and Al and between Zn and Al suggests that most of the Se and Zn in the surface sediment in the Western Harbour of Alexandria was anthropogenic in origin.

The correlation between Fe and Al is weak and many samples had high Fe concentrations. The high concentrations of Fe in the study area could be attributed to the presence of tens of floating rusty stranded barges. These barges could be major sources for particulate Fe that settles to the bottom sediments. The precipitated Fe in the form of oxhydroxide has affinity to scavenge other metals such as Cu, Zn and Pb as they pass through the water in route to the sediments (Waldichuk, 1985).

Compared to the results of a previous study (Table 2), concentrations of metals reported in the present study were several magnitudes higher than the metal levels reported for the same region in a previous study (El Sayed *et al.*, 1988). The large increase in the heavy metal concentrations in the surface sediment from the Western Harbour during the past decade was probably the consequence of the heavy industrial activities and urbanization in Alexandria. Continued discharge from agricultural, residential and industrial effluents to the harbor, and shipping activities were probably the major sources of anthropogenic heavy metals to the sediment. However, the samples from our study and the previous study might be collected from different sites and intervals in addition to different analytical method, which could contribute to some of the large differences in the metal concentrations. Compared to several estuaries in other parts of the world, the sediments in this study area have higher concentrations of metals than sediments from other contaminated harbours of the world (Table 2).

Possible Biological Effects

Many heavy metals are highly toxic and have chronic effects on living organisms. Elevated concentrations of heavy metals in sediments could cause detrimental effects to benthic organisms as well as other aquatic organisms. Most of the sediment samples from the Western Harbour of Alexandria appear to have heavy metal concentrations above the threshold that were deemed to be safe to benthic organisms (Table 3). Concentrations of Cd in majority of samples were below the Effects-Range Low (ERL) of Long and Morgan (1990) and Long *et al.* (1995), suggesting that most of the sediment samples may not have adverse effects on the benthic organisms (Table 3).

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Table 2

Comparison of some metal concentrations in sediments from Western Harbour of Alexandria and other harbors of the world ($\mu g\ g^{-1}$ dry weight)

Location	Al	Cd	Cu	Cr	Fe	Mn	Ni	Zn	Pb	References
Western Harbour, Alexandria, Egypt	9873–59204	0.61–2.44	39–207	33–649	8819–36140	139–317	13–53	58.5–382	38–1070	This study
Western Harbour, Alexandria, Egypt	NA	0.07–0.64	0.3–19	NA	5000–29000	15–40	NA	0.23–4.74	NA	El-Sayed *et al.*, 1988
Pasajes Harbour, Spain	NA	1.2–6.6.4	25–3726	NA	3830–35500	64–365	17–99	477–1390	45–346	Legorburu & Canton, 1991
Darwin Harbour, Australia	NA	0.9–3.1	3.7–33	NA	NA	NA	NA	18.3–270	22–91	Peerzada & Rohoza, 1989
Cork Harbour, Ireland	NA	<0.1	9–13	6–11	NA	NA	11–13	59–75	14–23	Berrow, 1991
Portsmouth Harbour, U.K.	NA	0.5–3.3	26–72	NA	NA	NA	NA	0.16–210	49–114	Soulsby *et al.*, 1978
Boston Harbour, USA	47300–82000	NA	7–142	42–292	9000–42000	NA	8–191	39–414	18–263	Bothner *et al.*, 2001

NA, No data available.

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Table 3
Number of samples that had metal concentrations above the sediment effects data of ERL and ERM in the Western Harbour of Alexandria. ERL and ERM guidelines were from Long and Morgan (1990) and Long *et al.* (1995). The ERL and ERM delineate concentrations at which adverse biological effects occur rarely (<ERL, <10% occurrence), occasionally (between ERL and ERM, 10–50% occurrence), and frequently (>ERM, >50% occurrence)

	As	Cd	Cr	Cu	Ni	Pb	Zn
Below ERL	7	11	10	5	13	4	8
Between ERL and ERM	14	10	9	15	7	13	10
Above ERM	0	0	2	1	1	4	3
ERL	8.2	1.2	81	34	20.9	46.7	150
ERM	70	9.6	370	270	51.6	218	410

Despite of the strong correlation between Al and Ni, many sediment samples had Ni concentrations above the ERL, indicating possible detrimental effects to benthic organisms in Alexandria Harbor. Similarly, most of the samples had Pb concentration above ERL and a few samples from Arsenal Basin, El-Mahmoudiya Quay and Mooring Area had Pb concentration above the Effects-Range Median (ERM) of Long and Morgan (1990) that is deemed to be detrimental to benthic organisms (Table 3). With regard to arsenic concentrations in sediment samples, they exhibited a range from 4.72 to 14.97 $\mu g\ g^{-1}$. The majority of these samples had As concentrations above the ERL.

Concentrations of chromium in samples from station 8 (649 $\mu g\ g^{-1}$) in the inner harbor and from station 20 (371 $\mu g\ g^{-1}$, near the outlet of the harbor) were above the ERM of Long and Morgan (1990), which indicate that the chromium would have significant detrimental effects on benthic organisms at these two locations. Many other samples had Cr concentrations above ERL, which could result in potential detrimental effects for benthic organism (Table 3).

Conclusions

Concentrations of many heavy metals were several times higher compared to the analysis conducted a decade ago in sediment of Western Harbour (e.g. 4 times for Cd, 11 times for Cu, 8 times for Mn and 75 times for Zn). This large increase in heavy metal concentration in the surface sediment was likely caused by the ever-increasing port and industrial activities during this period and continued discharge of agricultural and industrial effluents to the harbor. The higher levels of As, Cd, Cu, Mn, Ni, V, Sn, Cr, and Zn in the Arsenal Basin, and in the regions of the Boathouse, El Mahmoudiya outfall as well the Ship Lift, are mainly due to the presence of major sources of metal pollution and intensive human activities.

However, most metals yield significant linear correlation with Al, indicating that a major portion of the heavy metals in the sediment were closely associated with fine grained clay minerals and had common origins with Al. Some of the metals, such as As, Cr, Pb, Sn, and Zn, had poor correlation with Al. Anthropogenically sourced metals probably constituted a major portion for these metals in the sediment.

The concentrations of most heavy metals in the majority of the sediment samples were above the thresholds that were believed to be safe for living organisms. High levels of heavy metals could produce detrimental effects on benthic organisms. Although bioaccumulation

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of heavy metals was not investigated in this study, the high sediment concentrations of heavy metals in the Western Harbour of Alexandria could result in accumulation in biological system and produce adverse health effects.

Acknowledgment

We are grateful to Mr. K. El Nahta, Vice Chairman of the Port of Alexandria Corporation, for providing facilities during sample collection. Technical support by N. Eaker (Geochemical and Environmental Research Group, Texas A&M University) is appreciated. A.O.B. is grateful to the Fullbright Foundation for research fellowship. Constructive comments of the Managing Editor, D. Leonard, and other anonymous reviewers are highly appreciated.

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