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Heavy metal concentrations in surface sediments from the two coastal inlets (Golden Horn Estuary and İzmit Bay) of the northeastern Sea of Marmara

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ABSTRACT

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A total of 57 surficial sediment samples collected in the Golden Horn Estuary and İzmit Bay (northeastern Marmara Sea) was analyzed for the heavy metals Fe, Mn, Cr, Ni, Co, Zn, Pb, and Cu and the results were compared with various natural and anthropogenic sources.

It was observed that the concentrations of Zn, Cu, Pb, and, to some extent Cr, in the Golden Horn sediments are comparable with those in most other seas in the vicinity of highly industrialized and densely populated regions. Based on calculations from the metal:Al and metal:Fe ratios, the bottom sediments of Golden Horn were found to be enriched in Zn, Cu, Pb, and to a lesser extent Cr, by factors of up to 192 compared with the regional background levels of these metals. Fe, Mn, Co, and to some extent Ni, are mostly at natural levels. In contrast, the metal levels of the bottom sediments in İzmit Bay are significantly lower and they appear to be controlled by lithogenic rather than the anthropogenic influences.

Highly significant correlation coefficients between the metals Zn, Cu, and Pb are widely attributed to the common sources of these metals, the metal smelters and shipyards in the vicinity of the areas studied.

Based on the numerical value of the geoaccumulation index of heavy metals, it was found that the bottom sediments of İzmit Bay can be regarded as basically uncontaminated. Exceptionally high I_{geo} values are found for the bottom sediments of the Golden Horn, indicating moderate to extreme contamination in this region by the metals Pb, Zn, Cu, and Cr.

The presence of a large number of anthropogenic metal point sources, higher river run-off via the two major creeks, particularly coastal topography and hydrodynamic conditions, associated with the very high sedimentation rates, all strongly favour the accumulation of anomalously high metal concentrations in the Golden Horn Estuary, while the opposite is true for İzmit Bay.

1. Introduction

Coastal marine sediments from densely urbanized and industrialized regions have been subject to a good deal of research in the recent decades as they are the largest repository and potential source of metallic contaminants in the marine environment (e.g., Bruland et al., 1974; Thornton et al., 1975; Förstner, 1980; Ergin, 1990). Heavy metals, in particular, are amongst the best studied group of inorganic contaminants as metals from natural and anthropogenic sources accumulate together in sediments.

Although increasing contamination of the northeastern Marmara Sea, such as of the Golden Horn Estuary and the İzmit Bay, by heavy metals has been known for some time (e.g., Baykut, 1977; Taymáz et al., 1983; Orhon et al., 1984), up to now, no results of heavy metal geochemistry for sediments in these regions comparable to those presented here are available. Thus, the present work is aimed to fill a gap in our knowledge on the quantitative approximation of the heavy metal concentrations of northeastern Marmara sediments with respect to geological and anthropogenic sources.

2. The environmental setting

Figure 1 illustrates the two areas studied, which are located on the northeastern shelf of the Sea of Marmara, a transitional sea connecting the world's largest anoxic basin (the Black Sea) in the north to the evaporitic Mediterranean Basin in the south.

The Golden Horn Estuary (also called "İstanbul Haliç'i") is situated as a westerly tributary to the Strait of Bosphorus. It is approximately 7 km long and 150–900 m wide (on avg. 370 m) covering a surface area of about 2.6 km^2, having depths ranging from, approximately, 42 m at the mouth to 1 m at its upper end. The two creeks, Alibey and Kağıthane at present, discharge about 5×10^6 m^3/yr into the estuary. The İzmit Bay is located in the easternmost section of the Sea of Marmara. The bay is 50 km long and 2–10 km wide with a surface area of about 310 km^2. Water depths are around 25 m in the eastern part, 50–150 m in the centre and 50–200 m in the western part of the bay. In contrast to the Golden Horn, no major rivers enter the İzmit Bay.

The geological evolution of the Golden Horn

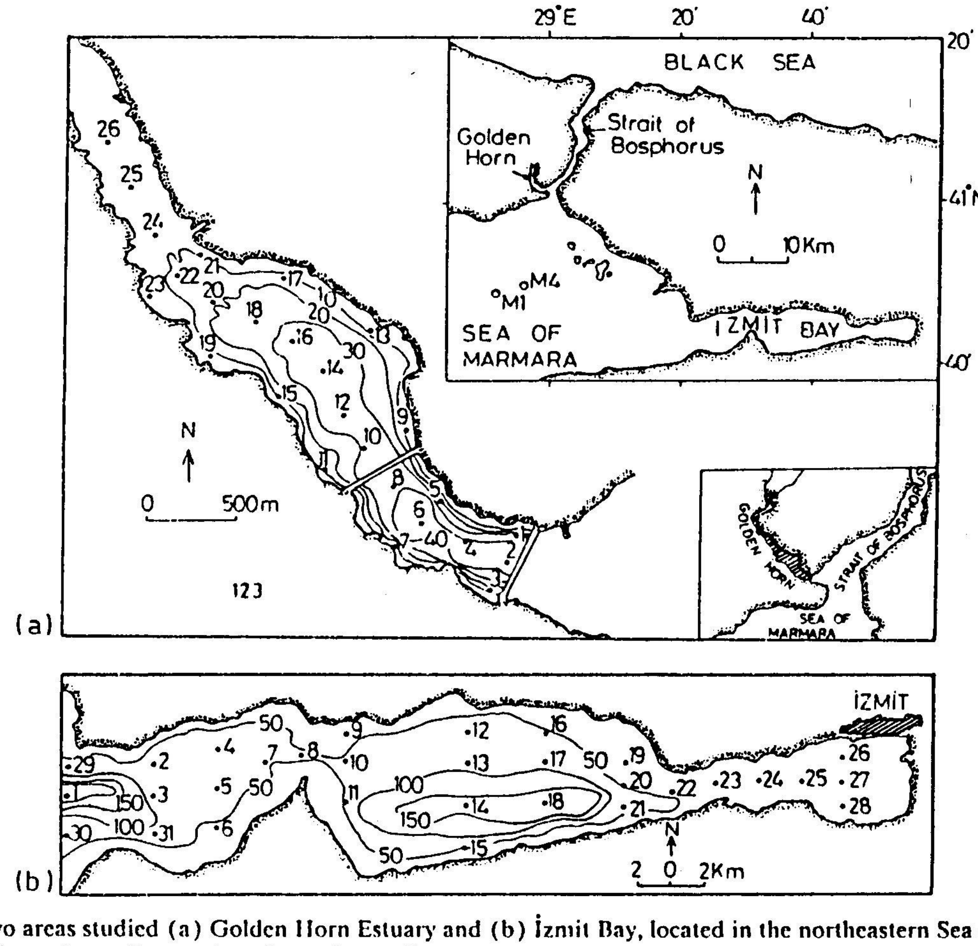

Fig. 1. The two areas studied (a) Golden Horn Estuary and (b) İzmit Bay, located in the northeastern Sea of Marmara with the locations of sampling stations for surface sediments.

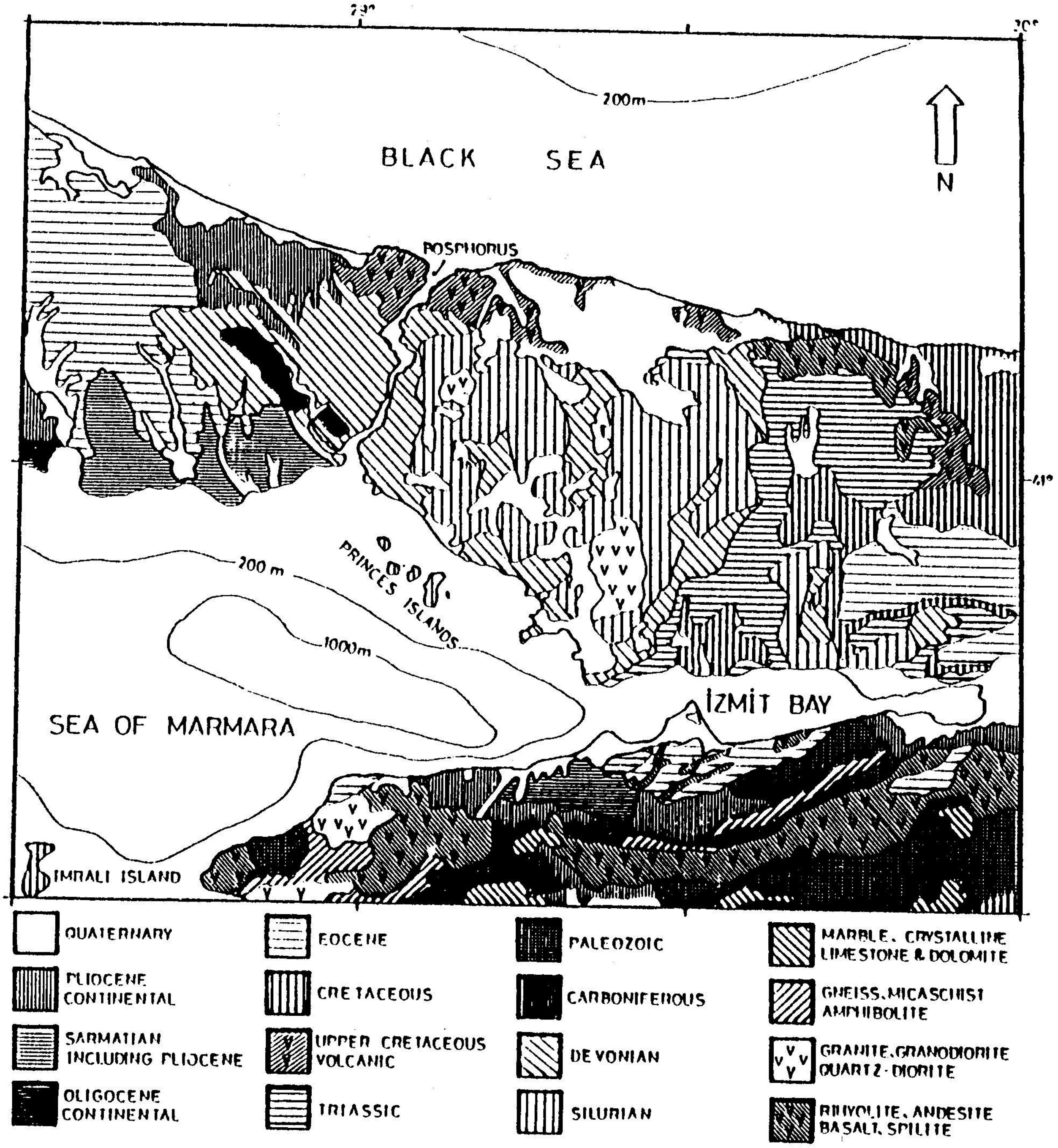

Fig. 2. Geology of the northeastern Marmara region (compilation map by Ternek, 1964).

Estuary and İzmit Bay is related to the late Tertiary epeirogenic crustal movements, which dissected and faulted the older structure to form graben-like depressions and valleys along the North Anatolian Fault Zone. Subsidence and transgressions during the Miocene/Pliocene or much later must have shaped most of their morphology (cf., Ergin and Yörük, 1990; Ergin et al., 1990). Figure 2 shows the major geological formations around the study area. In

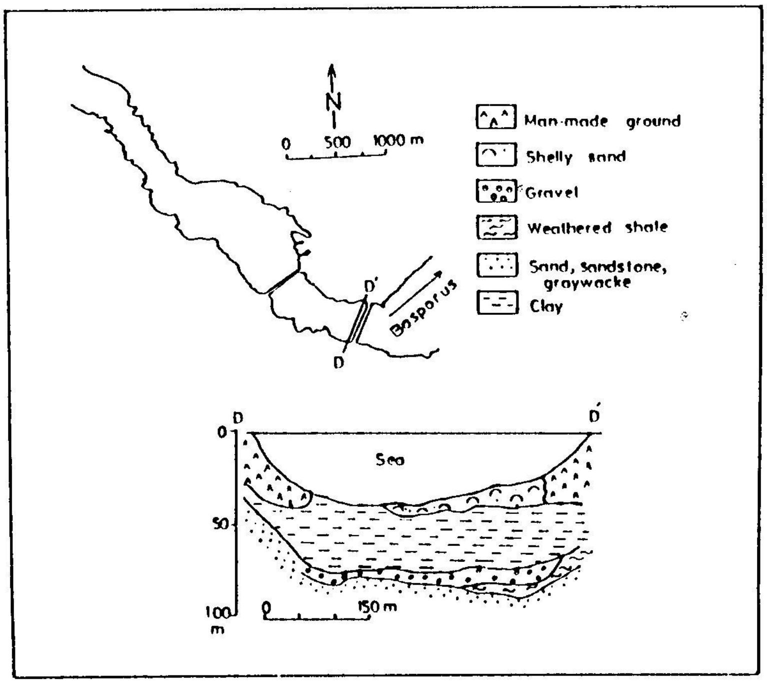

Fig. 3. A typical lithologic cross-section from the lower reach of Golden Horn Estuary (Özbek, 1990, personal communication).

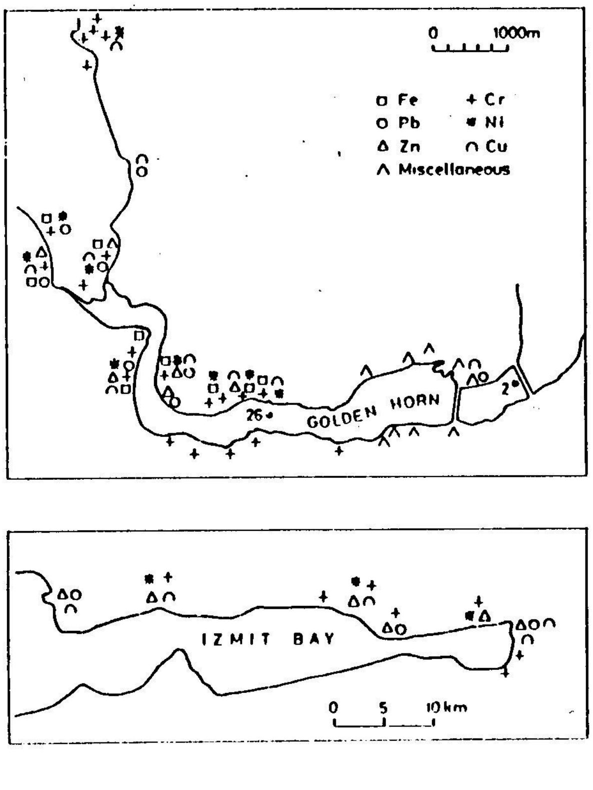

Fig. 4. The important discharge points for the heavy metals studied from anthropogenic sources (compiled from Karpuzcu, 1975 and Orhon et al., 1984). Note the large number of metal sources from the anthropogenic activities around the Golden Horn Estuary.

the Golden Horn, the older geological formations, now partly exposed on land, include graywackes, sandstones, and shales of late Paleozoic age (Fig. 3). The late Paleozoic is represented in the İzmit Bay by arkose, graywacke, marl, limestones, sandstones and schist.

The hydrologic regimes of the İzmit Bay and the Golden Horn Estuary are marked by a two-layered system of flows involving both the upper Black Sea (18–21 p.p.t. salinity) and the lower Mediterranean (38–39 p.p.t. salinity) waters (Baştürk et al., 1985; Saydam et al., 1986).

Various kinds of industries surround the Golden Horn (Baykut, 1977) and the İzmit Bay (Orhon et al., 1984) and discharge their wastes directly or indirectly into the system.

Compared to the İzmit Bay, the Golden Horn Estuary receives much more metal from the nearby anthropogenic sources (Fig. 4).

3. Materials and methods

Twenty-six samples of surface sediments were collected using a Van Veen® type of grab device in the Golden Horn Estuary in 1986 (Fig. 1). In the İzmit Bay, surface sediment sampling was conducted at thirty-one stations using a Dietz LaFonde® type of grab sampler in 1987 (Fig. 1). Bulk sediment samples from both areas were dried at approx. 50–60°C, powdered and throughly homogenized. Approximately 200–500 mg of these powdered samples was digested with HNO_3 acid on a sand bed for 2 h at 150°C. The leachable fractions of Fe, Mn, Cr, Ni, Co, Zn, Pb and Cu were then analyzed by an atomic absorption spectrophotometer (Varian® AA-*6* Model), some in duplicate and triplicate. The precision and accuracy of the analyses were checked with the international standards, such as CRM *142* (light sandy soil) and CRM *144* (sewage sludge of domestic origin) from the Community Bureau of Reference Materials (BCR), and the results are summarized in Table I. The analytical precision was normally between 1% and 11% for CRM *142* and between 1% and 20% for CRM *144*.

TABLE I

Accuracy of the method using two standards from the BCR (CRM *142* and *144*)

	CRM *142*		CRM *144*	
Cr (ppm)	44.4	(48)	494	(417)
Cu (ppm)	25.3	(28)	694	(567)
Mn (ppm)	527	(531)	436	(474)
Ni (ppm)	28.9	(27)	947	(725)
Pb (ppm)	30.9	(38)	479	(523)
Zn (ppm)	79.6	(76)	3090	–
Co (ppm)	7.9	(8)	9	(9.7)
Fe (%)	1.95	(1.78)	4.43	(4.71)

Values in parentheses were measured by this study (S. Yemenicioğlu and F. Yücesoy, personal communication). See text for reproducibilities.

For a quantitative measure of possible contamination in the studied sediments, the following equation introduced as the "Index of Geoaccumulation" (Müller, 1979) is used:

$$I_{geo} = \mathrm{Log}_2 \frac{C_n}{1.5 \times B_r}$$

where: C_n = the measured concentration; B_n = the geochemical background value; and 1.5 = a factor for lithologic variations of the heavy metal studied.

This method has successfully been used to determine the intensity of pollution by heavy metals in a given environment (Pons et al., 1988). In this work, two possibilities for establishing the background levels of the heavy metals will be discussed: average shale composition (after Turekian and Wedepohl, 1961), and metal levels from the deeper core sediments in the Golden Horn (see Fig. 3 for location) and the outermost İzmit Bay areas.

4. Results and discussion

The details of sediment characteristics, such as grain size, organic carbon and carbonate distribution are given elsewhere (Ergin and Yörük, 1990; Ergin et al., 1990). Bottom sediments in the İzmit Bay are dominated by mud, with varying proportions of clay (2–43%) and silt (12–94%). Total carbonate contents were between 2% and 45% $CaCO_3$. Organic carbon concentrations varied, on a carbonate-free basis, from 0.48% to 1.68%. In contrast, the floor of Golden Horn is covered with black, anoxic mud, where organic carbon contents ranged from 3.60–6.23%, calculated on a carbonate-free basis (9–15% $CaCO_3$). Microscopic studies reveal that, except for biogenic carbonates, the predominant siliceous clastic detritus of the İzmit Bay and Golden Horn sediments are comparable with the possible weathering products of the geological sources surrounding the study areas.

4.1. Distribution of heavy metals

The results of analyses of heavy metals in surface sediments from the Golden Horn Estuary and the Bay of İzmit are calculated for carbonate-free material and given in Tables II and III.

The heavy metal data show that the concentrations of Pb, Zn, Cu, Cr, and to lesser degree Ni, in the surface sediments of the Golden Horn are anomalously high compared to those found in the İzmit Bay. For example, the concentrations of Ni, Cr, Cu, Zn, and Pb in the Golden Horn sediments were up to 3, 40, 80, 76 and 14 times higher, respectively, than those from İzmit Bay. No major local metal anomalies are expected from the geochemical sources of the two regions under investigation. This is also apparent from the comparison with metal data from other areas in the northeastern Marmara Sea (Table IV). Apart from this, sediment investigations from the boreholes drilled in the lower Golden Horn (40 m water depth) revealed that the concentrations of Pb, Zn, Cu, Cr, and Ni, from depths between 10 and 35 below the sea floor were remarkably low and uniform, reflecting most probably the natural levels of these metals in this region (Özbek, 1990, personal communication). As can be inferred from Tables II and IV, the sediments of the Golden Horn are enriched in metals Cu, Zn, Pb, Cr, and Ni by factors of 105, 94, 28, 3, and 2, respectively, when compared with the natural background levels of this estuary. On the other hand, comparison of the heavy metal

TABLE II

Concentrations of heavy metals in surface sediments from the Golden Horn

Sample	Pb (ppm)	Zn (ppm)	Cu (ppm)	Cr (ppm)	Co (ppm)	Ni (ppm)	Mn (ppm)	Fe (wt%)	Al (wt%)
H-*1*	274	916	676	339	19	111	426	3.7	5.7
H-*2*	261	682	608	308	19	113	407	3.5	6.1
H-*3*	402	930	1534	323	16	136	454	3.3	6.0
H-*4*	385	2168	1318	372	26	143	427	3.6	6.0
H-*5*	797	9943	4432	355	22	189	432	3.5	4.8
H-*6*	244	831	676	275	19	111	406	3.3	6.1
H-*7*	389	1029	1148	339	26	133	426	3.5	5.4
H-*8*	291	909	642	308	22	120	378	3.5	5.4
H-*9*	382	1903	1170	372	23	133	384	3.3	5.4
H-*10*	287	1022	800	324	23	124	393	3.4	4.8
H-*11*	445	1058	808	339	19	168	458	2.9	2.6
H-*12*	270	966	811	372	26	139	429	3.3	6.6
H-*13*	274	979	586	291	33	136	452	3.8	6.0
H-*14*	141	546	620	291	29	117	421	3.7	6.1
H-*15*	402	566	337	323	26	95	485	4.0	5.8
H-*16*	317	1243	1103	437	29	129	427	3.5	6.5
H-*17*	368	1377	1250	485	26	152	409	3.7	6.3
H-*18*	364	1711	1440	502	33	159	432	3.7	6.3
H-*19*	368	1562	1453	453	26	175	407	3.8	6.5
H-*20*	249	894	1081	372	23	127	412	3.7	6.3
H-*21*	300	1079	642	420	26	155	386	4.1	6.2
H-*22*	141	511	912	372	32	148	642	3.4	7.1
H-*23*	578	1412	1025	453	33	177	518	4.3	6.0
H-*24*	300	1228	1250	469	29	177	525	4.1	–
H-*25*	286	1001	1241	551	33	171	563	3.9	6.0
H-*26*	143	724	1114	420	35	152	642	3.6	7.5

Al contents from Evans (1986, personal communication). All results are on a carbonate-free basis.

TABLE III

Concentrations of heavy metals in surface sediments from İzmit Bay

Sample	Pb (ppm)	Zn (ppm)	Cu (ppm)	Cr (ppm)	Co (ppm)	Ni (ppm)	Mn (ppm)	Fe (wt%)
1	53	83	27	35	93	77	193	3.47
2	51	94	35	31	66	95	216	2.76
3	45	92	25	27	116	82	179	2.76
4	43	76	26	25	111	93	171	2.53
5	38	68	23	7	111	93	171	2.53
6	24	75	26	66	66	80	149	2.64
7	42	65	24	32	89	53	240	1.77
8	38	62	18	46	86	46	222	1.92
9	59	128	29	28	91	90	159	2.57
10	83	114	34	45	123	154	349	3.23
11	39	94	27	27	59	56	167	2.74
12	49	94	27	28	96	58	244	2.65
13	47	89	29	61	80	90	291	3.10
14	39	91	30	34	62	67	745	2.91
15	39	77	24	19	63	49	198	3.03
16	42	85	27	34	94	90	199	3.62
17	48	89	24	89	74	105	287	3.52
18	40	92	31	14	103	91	-	2.72
19	31	75	21	21	94	48	127	4.45
20	36	83	26	14	77	104	139	3.54
21	30	84	29	60	65	59	207	3.11
22	28	47	18	-	43	59	138	1.38
23	31	76	26	16	115	54	176	3.02
24	36	70	30	40	76	47	133	4.17
25	36	78	34	52	63	72	145	3.62
26	35	92	51	40	98	90	133	4.00
27	29	77	40	39	85	43	155	2.35
28	25	60	33	25	64	45	192	3.70
29	39	83	24	35	73	105	156	3.13
30	41	86	28	34	60	71	163	2.77
31	47	83	23	34	54	71	147	3.13

All results are on a carbonate-free basis.

TABLE IV

Comparison of the background levels of heavy metals from the northeastern Marmara Sea with average shale composition

	Average shale[1]	Golden Horn[2]	Marmara/Bosphorus junction[3]	Outermost İzmit Bay[4]
Pb (ppm)	20	28	35	45
Zn (ppm)	95	106	82	107
Cu (ppm)	45	42	28	49
Cr (ppm)	90	164	83	138
Co (ppm)	19	32	9	30
Ni (ppm)	68	113	60	107
Fe (wt%)	4.7	5.2	2.5	4.9
Mn (ppm)	850	524	470	1256[5]
Al (wt%)	8.0	7.6	-	7.9
$CaCO_3$ (wt%)	-	15	15	17

[1]Turekian and Wedepohl (1961); [2]Özbek (personal communication, 1990); [3]Bodur (personal communication, 1990); [4]Ergin and Evans (1988) and Evans et al. (1989); [5]Orhon (1984).

TABLE V

Heavy metal concentrations from various sources

Location	Pb (ppm)	Zn (ppm)	Cu (ppm)	Cr (ppm)	Co (ppm)	Ni (ppm)	Mn (ppm)	Fe (ppm)	Al (ppm)
This study									
[1]İzmit Bay	23- 52	45- 114	13- 49	6- 81	43-105	34- 98	112- 678	1.40-3.97	-
[1]Golden Horn Estuary	124-702	450-8750	333-3900	242-485	17- 31	98-167	333- 565	2.60-3.80	2.3 -6.6
Coastal areas of Turkey									
[2]North, off Bosphorus	15- 48	42- 101	16- 54	13-169	<15	3- 75	123- 450	0.29-3.30	-
[3]South, off Bosphorus	34- 51	85- 97	38- 40	111-124	8- 16	53- 60	360- 388	2.49-2.91	-
[4]Eastern Marmara Basin	52	102	46	118	24	87	500	4.05	6.5
[5]Southwestern Black Sea	22	129	24	193	21	79	832	4.50	7.5
[6]Eastern Aegean	17	39	18	92	16	143	925	2.41	0.9 -8.5
[7]Mersin Bay (NE-Mediterranean)	7- 63	25- 95	8- 65	18-611	11- 40	152-428	236-1716	1.10-5.10	4.70
[8]Mersin Harbour	144	250	136	-	-	266	457	3.8	-
[9]İzmir Bay, Turkey	40-280	53- 860	14- 866	-	-	-	-	1.3 -4.4	-
Other areas of the World									
[10]Thermaikos Gulf, Greece	45-310	80-2400	35- 70	80-280	15- 35	90-440	650-2600	3.5 -5.5	-
[11]Gulf of Venice, Italy	5- 84	48- 870	3- 44	10-254	1- 14	5- 41	-	0.7 -6.7	-
[12]Rio Tinto Estuary, Spain	1600	3100	1400	-	-	-	-	-	-
[13]Weser Estuary, Germany	25-142	59- 377	23- 28	-	-	22- 24	307-1402	2.95-4.25	2.96-3.46
[14]Eckernförder Bay, Germany	102	272	42	38	9	41	500	4.0	6.0
[15]Ganges Estuary, India	12-115	12- 611	4- 53	21-100	14- 64	8- 57	254- 800	1.2 -4.6	3.6 -8.0
[16]Derwent Estuary, Australia	1000	10000	-	258	-	-	-	-	-
[17]Restonguet Estuary, U.K.	1620	3000	4500	1060	-	-	-	-	-
Average sedimentary rocks									
[18]Average shale	20	95	45	90	19	68	850	4.7	8.0
[18]Average sandstone	7	16	10	35	<1	2	50	0.98	2.5
[18]Average carbonate	9	20	4	11	<1	20	1100	0.38	0.4
Deeper core sediments, adjacent to the Sea of Marmara									
[19]Eastern Marmara	38	89	41	115	25	89	265	4.1	6.6
[20]Southwestern Black Sea	27	128	29	185	18	84	672	4.6	7.7
[21]Golden Horn	28	106	42	164	32	113	524	5.2	7.6
[22]South, off Bosphorus	35	82	28	83	9	60	470	2.5	-

[1]This study; [2]Yücesoy (personal communication, 1990); [3]Bodur (personal communication, 1990); [4]Ergin and Evans (1988); Evans et al. (1989); [5]Hirst (1974); [6]Voutsinou-Taliadouri and Satsmadjis (1982); [7]Shaw and Bush (1978); [8]Özkan et al. (1980); [9]Uysal and Tuncer (1984); [10]Voutsinou-Taliadouri (1982); [11]Donazzolo et al. (1981); [12]Stenner and Nickless (1975); [13]Shoer et al. (1982); [14]Ergin (1990); [15]Subramanian et al. (1988); [16]Bloom and Ayling (1977); [17]Thornton et al. (1975); [18]Turekian and Wedepohl (1961); [19]Ergin and Evans (1988); Evans et al. (1989); [20]Hirst (1974); [21]Özbek (personal communication, 1990); [22]Bodur (personal communication, 1990).

data in this study with those from other coastal environments in many parts of the world (Table V) indicates a wide agreement that the Golden Horn estuary seems to be comparable with most other seas in the vicinity of highly industrialized and densely populated regions.

In contrast, the metal levels of the İzmit Bay sediments, with the exception of Fe and Co, are significantly lower than the Golden Horn values (Table V). Fe concentrations appear to show a similar distribution in the two study areas. The Co concentrations in the İzmit Bay sediments are three times higher, and the Mn levels are two times lower than those found in the Golden Horn. The differences in the distribution patterns of Co and Mn are, most probably, due to post-depositional processes, which might be different in the two inlets. Mn and Co are known to be the mobile elements most sensitive to redox changes in the depositional environment (e.g., Manheim, 1965; Delfino and Lee, 1968; Elderfield, 1977).

4.2. Enrichment factors for the metals

The overall approach taken in this study was to compare the measured heavy metal concentrations of sediments by acquiring available, compatible data from a variety of natural sources, which in turn, are believed to be representative of "geochemical" or "pre-industrial" materials in the northeast Marmara region. These include, apart from the average shale standard, baseline levels of metals in deeper core sediments from the Golden Horn, the outermost İzmit Bay, and the Sea of Marmara/Bosphorus junction (Table IV). Average shale composition is a global reference basis and satisfies the basic requirement of being a source of uncontaminated fossil argillaceous sediment, when no local background values are available (e.g., Förstner and Müller, 1974).

To reduce the metal variability caused by grain size and mineralogy, and to identify anomalous metal contributions, geochemical normalization of the heavy metal data to "conservative" elements, such as Al (Kemp et al., 1976), and Fe (Rule, 1986) has been used in this study. The resulting Metal/Al ratios would then be suggestive of the metal enrichment factors, *EF*:

$$EF_{(Me)} = \frac{(conc_{Me}/conc_{Al\ or\ Fe})_{sample}}{(conc_{Me}/conc_{Al\ or\ Fe})_{background}}$$

Metal/Al and Metal/Fe ratios for the sediment samples studied and some others are given in Table VI, and the calculated enrichment factors thereof are listed in Table VII, for comparison. It is obvious from Table IV, that the background levels of heavy metals from deeper core sediments (carbonate-free basis) in the Golden Horn, Bosphorus–Marmara junction, and the outermost İzmit Bay areas appear to be generally comparable to the average shale composition, and the variations must primarily be related to the changes in the lithology.

In contrast to the sediments of İzmit Bay, those from the Golden Horn Estuary exhibit

TABLE VI

Comparisons of Me/Al and Me/Fe ratios in sediments of this study with those from various sources (see text for details)

Ratio	Golden Horn (this study)	İzmit Bay (this study)	Outermost İzmit Bay[2]	Golden Horn[3]	Black Sea[4]	Average shale[5]	Diagenous Black Sea[6]
Fe/Al	0.63– 0.96	n.d.	0.6	0.7	0.6	0.58	0.8
Pb/Al ($\times 10^{-4}$)	16 – 153	n.d.	6	3.7	4	2	3
Pb/Fe ($\times 10^{-4}$)	41 – 226	8– 23	9	5.4	5	4	(6.3)
Zn/Al ($\times 10^{-4}$)	84 –2302	n.d.	13	14	16	12	14
Zn/Fe ($\times 10^{-4}$)	150 –2822	16– 51	22	20	26	20	(26)
Cu/Al ($\times 10^{-4}$)	55 – 661	n.d.	6	5.5	4	6	5
Cu/Fe ($\times 10^{-4}$)	82 –1258	7– 13	10	8.1	6	9	5.8
Cr/Al ($\times 10^{-4}$)	31 – 110	n.d.	17	21	24	11	7
Cr/Fe ($\times 10^{-4}$)	77 – 175	1– 25	28	31	37	19	9
Co/Al ($\times 10^{-4}$)	4	n.d.	4	4.2	2	2	2
Co/Fe ($\times 10^{-4}$)	5 – 10	17– 42	6	6.1	4	4	3
Ni/Al ($\times 10^{-4}$)	20 – 28	n.d.	13	14.8	11	9	6
Ni/Fe ($\times 10^{-4}$)	23 – 57	11– 43	22	21.7	17	14	8
Mn/Al ($\times 10^{-4}$)	67 – 96	n.d.	39	69	87	106	566
Mn/Fe ($\times 10^{-4}$)	107 – 188	64–255	64	100	134	181	685

[2]Ergin and Evans (1988); Evans et al. (1989); [3]Özbek, (personal communication, 1990); [4]Hirst (1974); [5]Turekian and Wedepohl (1961); [6]Sevast'yanov and Volkov (1967).
n.d. = not determined.

TABLE VII

Enrichment factors of heavy metals in sediments from the Golden Horn Estuary and İzmit Bay, together with data from various sources for comparison (see text for details)

	Golden Horn (this study)		İzmit Bay (this study)		Black Sea sediments (diagenous)[1]		Red Sea sediments (hydrogenous)[2]	
Zn/Al	7–192	(6–158)			1.2	(0.9)	14 -3900	(12 -3172)
Zn/Fe	7–141	(6–117)	<1– 3	(<1–2)	1.3	(1.1)	1 - 95	(0.6– 79)
Pb/Al	8– 77	(3– 31)			1.5	(0.6)	260 -1100	(104 - 440)
Pb/Fe	10– 56	(6– 32)	2– 6	(1–3)	1.6	(0.9)	1 - 13	(0.3– 7)
Cu/Al	9–110	(11–132)			0.8	(1)	185 -3000	(220 -3600)
Cu/Fe	9–140	(10–157)	<1– 2	(<1–2)	0.6	(0.7)	4 - 30	(5 - 34)
Cr/Al	3– 10	(1– 5)			0.6	(0.3)	0.2– 4	(0.1– 2)
Cr/Fe	4– 9	(2– 5)	<1	(<1)	0.5	(0.1)	0.1– 1.2	(0.1– 0.7)
Ni/Al	2– 3	(1– 3)			0.7	(0.5)	0.2– 1.1	(0.1– 0.8)
Ni/Fe	2– 4	(1– 3)	<1– 4	(<1–2)	0.6	(0.4)	0.1– 0.5	(0.1– 0.4)
Co/Al	2	(2)			1	(0.6)	7 - 150	(5 - 100)
Co/Fe	1– 3	(1– 2)	4–11	(3–9)	0.8	(0.6)	0.5– 2	(0.4– 1.6)
Mn/Al	<1	(1– 2)			5	(9)	5 - 75	(8.7– 127)
Mn/Fe	<1	(1– 2)	<1	(<1–3)	4	(7.3)	0.3– 0.8	(0.5– 1.7)
Fe/Al	1– 2	(1– 2)			1.4	(1.3)	2 - 86	(1.6– 82)

[1]Sevast'yanov and Volkov (1967); [2]Hendricks et al. (1969).
Values in parentheses are based on average shale composition.

the highest enrichment factors for Zn, Pb, and Cu, with values up to 200. Based on the above observations, the results of the petrography of the sediments studied, and the geology of the surrounding land masses, it is more likely that the elevated levels of heavy metal concentrations, particularly those in the Golden Horn estuary, can not be explained by the lithogenic sources.

However, as is known, the heavy metal anomalies in Recent marine sediments may also be derived, apart from the increased human activities, from other natural/geochemical processes, such as diagenesis (e.g., Sevast'yanov and Volkov, 1967; Delfino and Lee, 1968; Elderfield and Hepworth, 1975; Calvert and Price, 1977; Ridgway and Price, 1987; Loring and Rantala, 1988; Szefer, 1988), submarine hydrothermal/volcanic activities (Hendricks et al., 1969; Cronan, 1977), and weathering of particular metal-rich deposits on land (Rose et al., 1979). The latter possibility is well documented for the sediments from Mersin Bay (northeast Mediterranean), where mafic–ultramafic rocks from the coastal hinterland supply high background concentrations, particularly for the metals Cr (18–611 ppm and Ni (1952–753 ppm) (Shaw and Bush, 1978; Bodur and Ergin, 1988). Petrographic investigations of the sediment samples, however, reveal no important sources of the metals in the vicinity of Golden Horn and İzmit Bay.

Hydrothermal activity might have been expected in the study area because the Sea of Marmara is located along the North Anatolian Fault Zone that has, in recent times, been tectonically active, mostly in form of earthquakes (Brinkman, 1976; Ketin, 1983). However, no heavy metal deposits related to such submarine activity are known from the study area (in contrast to occurrences in the Red Sea). In the Red Sea, for example, the recent metal deposits of hydrothermal origin display a very different pattern of metal distribution; metal enrichment factors (up to 4000) on the basis of Me/Al and Me/Fe ratios are considerably higher than those found in this study (up to 200) (Table VII).

The role of diagenetic processes could also be considered as an additional mechanism for the accumulation of high metal concentrations, especially in the Golden Horn sediments. This is due to the fact that the anoxic bottom sediments of the Golden Horn are temporarily subject to oxidizing conditions by the inflowing oxygen-rich undercurrents from the Sea of Marmara. However, the redox-sensitive elements such as manganese do not confirm any relevant diagenetic metal enrichment in the studied sediments, in contrast to the Black Sea sediments where Mn/Al and Mn/Fe ratios (566–685) are known to be much higher than those obtained in this study (67–188—Table VI).

4.3. Control, sources, and geoaccumulation indices of heavy metals in the two coastal environments

From the above discussions it appears that the heavy metal contents of the İzmit Bay sediments must be widely controlled by lithogenic influences, whilst the Golden Horn sediments reflect predominantly anthropogenic influences, although both of the studied coastal inlets are subject to the increased environmental pollution by heavy metals.

Figure 4 shows the approximate sites of waste discharges relatively rich in heavy metals from various domestic and industrial

TABLE VIII

The calculated geoaccumulation indices of heavy metals in the Golden Horn Estuary and their designation of sediment quality

Metal	Measured concentration (ppm)	Index of geoaccumulation*		Sediment quality
Fe	2.90– 4.30 %	<0	(<0)	uncontaminated
Mn	378 – 642	<0	(<0)	uncontaminated
Co	16 – 35	<0	(<0 –0.3)	uncontaminated/moderately contaminated
Ni	95 – 189	<0 –0.1	(<0 –0.9)	uncontaminated/moderately contaminated
Cr	275 – 551	0.2–1.2	(0.3–2.0)	moderately contaminated
Cu	337 –4432	2.4–6.1	(2.3–6.0)	moderately/extremely contaminated
Zn	511 –9943	1.7–6.0	(1.8–6.1)	moderately/extremely contaminated
Pb	141 – 797	1.7–4.2	(2.2–4.7)	moderately/strongly contaminated

*Values in parenthesis are based on average shale composition (Turekian and Wedepohl, 1961), whilst others are obtained from the deeper sections of core sediments (carbonate-free basis) in the Golden Horn Estuary, near station *2* (Özbeck, 1990, personal communication).

TABLE IX

The calculated geoaccumulation indices of heavy metals in İzmit Bay and their designation of sediment quality

Metal	Measured concentrations (ppm)	Index of geoaccumulation*		Sediment quality
Fe	1.38– 4.17 %	<0	(<0)	uncontaminated
Mn	127 –745	<0	(<0)	uncontaminated
Co	43 –123	<0–1.4	(0.6–2.1)	uncontaminated/moderately contaminated
Ni	43 –154	<0	(<0 –0.6)	uncontaminated/moderately contaminated
Cr	7 – 89	<0	(<0)	uncontaminated
Cu	18 – 51	<0	(<0)	uncontaminated
Zn	47 –128	<0	(<0)	uncontaminated
Pb	24 – 83	<0–0.3	(0 –0.2)	uncontaminated/moderately contaminated

*Values in parenthesis are based on average shale composition (Turekian and Wedepohl, 1961), whilst others are calculated from the deeper sections of sediment cores from the outermost İzmit Bay (Orhon, 1984; Ergin and Evans, 1988; Evans et al., 1989).

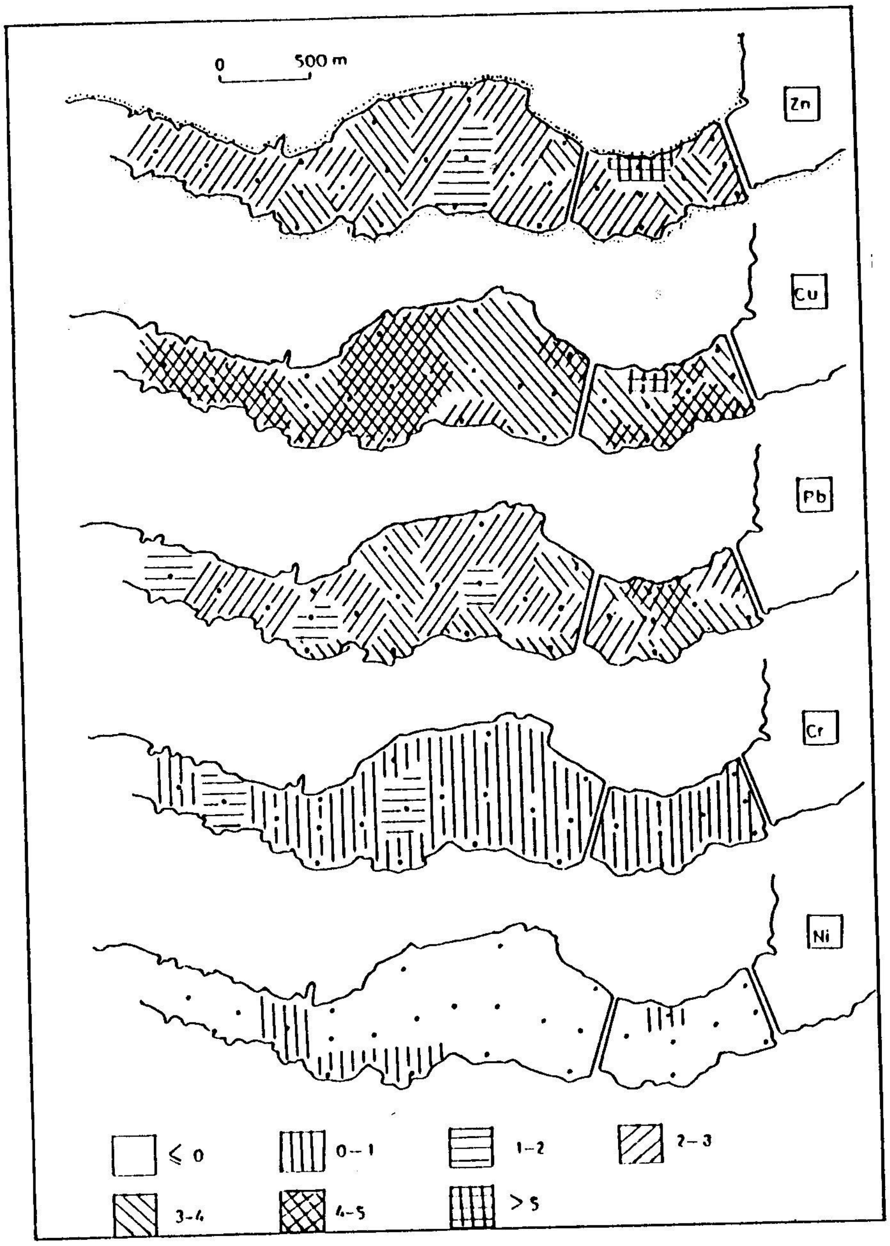

Fig. 5. Indices of geoaccumulation of the studied heavy metals in the Golden Horn surface sediments. Note the uncontaminated (< 0), moderately-contaminated (1–3), and extremely-contaminated (> 5) sediments by the heavy metals. See text as well as Table VIII for details.

sources. As a result, the Golden Horn sediments must receive much more anthropogenic metal input than do those of İzmit Bay.

Also, the hydrography and related water flows which are different in the two coastal environments should play an important role in the metal distribution. As mentioned earlier, İzmit Bay is much wider, deeper, and longer than the Golden Horn Estuary, resulting in movements of large volumes of water and, thus, great dilution for the heavy metals entering İzmit Bay. In addition, the outflowing Golden Horn waters are commonly trapped or blocked by the southerly flowing Bosphorus waters; a

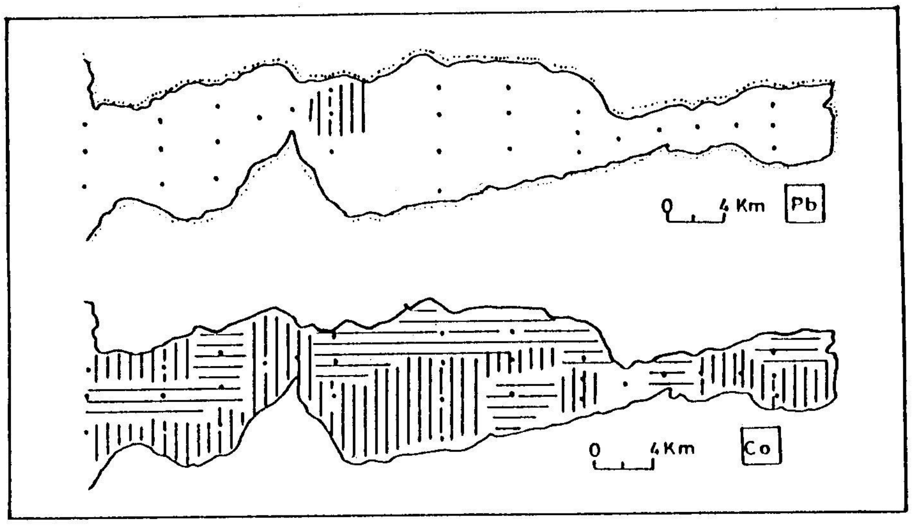

Fig. 6. Indecies of geoaccumulation of the studied heavy metals in İzmit Bay surface sediments. Note the almost uncontaminated sediments by the heavy metals. I_{geo} values for Co may come partly from the post-depositional mobilization in the sediment. See text as well as Table IX for details.

fact which causes longer residence times for the waters within the estuary.

The accumulation rates of the modern sediments in the Golden Horn estuary (6–10 cm/yr: cf Balcı, 1977) are much higher than those calculated for the İzmit Bay sediments (0.25 mm/yr: Ergin and Yörük, 1990). Thus, significantly higher amounts of heavy metal concentrations must be introduced into the bottom sediments of Golden Horn compared to those in İzmit Bay.

The indices of geoaccumulation (I_{geo}) of the heavy metals were obtained in this study and are given in Tables VIII and IV and the related regional distribution is presented in Figs. 5 and 6. Based on the numerical values of the indices, it is safe to say that the bottom sediments of İzmit Bay can be regarded as basically uncontaminated since I_{geo} values for all metals were usually less than 1, except for Co which had I_{geo} values up to 2 (Table IX; Fig. 6). Exceptionally high I_{geo} values were found for Zn and Cu (> 5) as well as for Pb (4–5) in the bottom sediments at Station 5, in the lower Golden Horn estuary (Table VIII; Fig. 5) suggesting similar anthropogenic sources and source points for these metals. The principal sources of Zn, Cu, and Pb are believed to be the small but numerous commercial metal smelters and shipyard activities in the vicinity. Cr and Ni values indicate no more than moderate contamination ($I_{geo}<2$; Table VIII) which must probably be caused by the Cr-consuming textile and leather tanning industries and the Ni-consuming electrometal industries, battery plants and dockyards.

4.4. Heavy metal associations within the sediment

Correlations between the heavy metals studied are statistically tested and the relationship between two such parameters are shown on a Pearson correlation coefficient matrix in Tables X and XI, for the İzmit Bay and Golden Horn Estuarine surface sediments, respectively.

As shown in Fig. 7, concentrations of Zn, Cu, and Pb in the Golden Horn sediments correlate strongly with each other ($r=+0.72$, $+0.77$, $+0.94$); whereas those of Zn with Cu and Pb are less significant in the İzmit Bay sediments (Table X: $r=+0.46$, $+0.58$). In particular, in the vicinity of the Golden Horn, these elements have similar anthropogenic

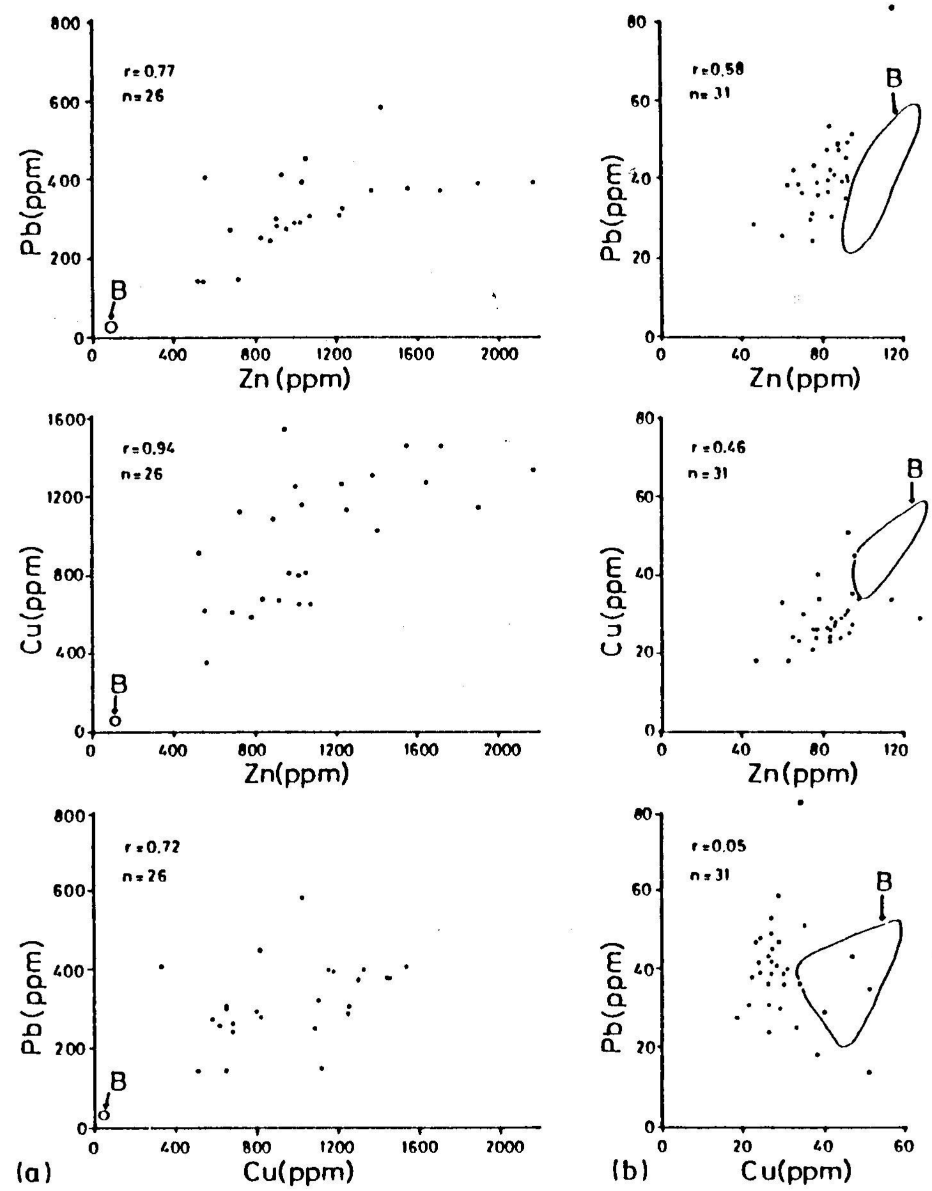

Fig. 7. Inter-element correlations for some metals in (a) the Golden Horn and (b) İzmit Bay surface sediments. Note the nearly natural metal levels in the İzmit Bay, largely consistent with the regional background (*B*) values, in contrast to those in the Golden Horn (see text for details).

sources, mainly represented by the metal smelters and shipyards. However, Zn, Cu, and Pb together may also occur in sediments associated with the "coal-residue assemblage", as shown by Erlenkeuser et al. (1974) and Ergin (1990). It thus appears that the presence of coal and slag particles in the samples studied from the Golden Horn could be indicative of such fossil fuel combustion and of the resulting release of Zn, Cu, and Pb to the estuary.

The Cr and Ni values of the Golden Horn sediments show good correlation with each other ($r = +0.66$; Table XI), although the anthropogenic sources of these metals are markedly different, as mentioned above. Obviously, the distribution of these metals is

TABLE X

Pearson correlation coefficient matrix for heavy metals in the surface sediments from İzmit Bay

	Fe	Mn	Co	Pb	Zn	Ni	Cr	Cu
Fe	1.00							
Mn	0.16	1.00						
Co	0.13	0.19	1.00					
Pb	0.10	0.07	0.09	1.00				
Zn	0.43	0.17	0.19	0.58	1.00			
Ni	0.33	0.16	0.17	0.48	0.44	1.00		
Cr	0.42	0.13	0.16	0.14	0.35	0.26	1.00	
Cu	0.64	0.09	0.17	0.05	0.46	0.14	0.32	1.00

$r=0.349$; 5% level of significance
$r=0.449$; 1% level of significance
$n=31$

TABLE XI

Pearson correlation coefficient matrix for heavy metals in the surface sediments from the Golden Horn Estuary

	Fe	Mn	Co	Pb	Zn	Ni	Cr	Cu
Fe	1.00							
Mn	0.17	1.00						
Co	0.52	0.61	1.00					
Pb	0.68	0.22	0.21	1.00				
Zn	0.31	0.13	0.11	0.77	1.00			
Ni	0.20	0.33	0.37	0.48	0.48	1.00		
Cr	0.43	0.33	0.55	0.11	0.04	0.66	1.00	
Cu	0.06	0.01	0.55	0.72	0.94	0.59	0.21	1.00

$r=0.380$; 5% level of significance
$r=0.486$; 1% level of significance
$n=26$

believed to be controlled by water circulation and modern sedimentation processes within the estuary (Ergin et al., 1990), rather than a source from outside. On the other hand, the significant correlations existing between Co and Cr, Mn, and Fe ($r=0.52$–0.66; Table VI) are probably suggestive of post-depositional processes within the sediment. The good correlation between Fe and Pb ($r=+0.68$) is considered to be due to contributions partly from the metal and electrolytic industries, or/and from the association of Pb with Fe compounds, such as iron oxides/hydroxides.

In the İzmit Bay, significant correlations exist between Cu with Fe ($r=+0.64$), and Zn ($r=+0.46$), as well as Pb with Zn ($r=+0.58$) and Ni ($r=+0.48$) (Table X). However, these observations again reflect lithogenic/post-depositional control on these metals because of the low metal concentrations at nearly natural levels.

5. Conclusions

On the basis of the observed lithological characteristics, and the comparisons of heavy metal concentrations obtained in this study with those from other contaminated and uncontaminated sedimentary materials, the following conclusions can be drawn:

(1) Golden Horn sediments contain remarkably high concentrations of Zn, Cu, Pb, and Cr when compared with those from the İzmit Bay.

(2) Based on the Me/Al and Me/Fe ratios, the concentrations of Zn, Cu, Pb, and Cr measured in the Golden Horn sediments were found to be enriched by factors of 3–192 when compared with the possible background levels of these metals in this region.

(3) Evidence from geo-accumulation indices showed moderate to extreme contamination in the Golden Horn Estuary by the metals Zn, Cu, Pb, and to lesser extent by Cr.

(4) Whilst the heavy metal concentrations of the İzmit Bay sediments would be largely consistent with the natural background levels, and thus, reflect the dominance of lithogenic influences, those of the Golden Horn indicate important anthropogenic influences from the nearby industrial activities.

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