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Biomonitoring study of heavy metals in biota and sediments in the South Eastern coast of Mediterranean sea, Egypt

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Abstract Concentrations of Cd, Cu, Co, Zn, Mn and Fe were determined in biota and sediment samples collected from the Eastern Harbour and El-Mex Bay in the Mediterranean Sea, Egypt. The levels of Cu, Co, Zn, Mn and Fe in the macroalgae, *Ulva lactuca*, *Enteromorpha compressa* (green algae) and *Jania rubens* (red algae), recorded high concentrations except for Cd. Moreover, Fe was the most predominant metal in the seaweed. The two species of bivalves, *Donax trunculus* and *Paphia textile,* showed different amounts of metals in their tissue. The abundance of heavy metal concentrations in the mussel samples was found in the order Fe> Zn> Mn> Cu> Co> Cd and Fe> Zn> Mn> Cu> Cd> Co, respectively for the two species. The metals concentrations were generally higher compared with the previous studies in mussels from the same area. The levels of metals accumulated in the investigated fish samples, *Saurida undosquamis*, *Siganus rivulatus*, *Lithognathus mormyrus* and *Sphyraena sphyraena*, were higher than those of Marmara Sea (Turkey), for Co and Cd and lower for Cu, Zn, Mn and Fe. El-Mex Bay having the highest metals concentration in sediments as their order of abundance were Fe> Zn> Mn> Cu> Cd> Co. Nevertheless, a high variability in the metal levels occurs among the studied algae and biota and also between the investigated Harbour. A significant correlations ($p<0.05$) were found for each of Zn and Fe in *P. textile* and of Co in *D. trunculus* relative to their concentrations in surficial sediments.

Keywords Algae · Biota · Heavy metals · El-Mex Bay · Eastern Harbour · Sediments

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Introduction

El-Mex Bay and the Eastern Harbour (E.H) are two parts of Alexandria coast on the Mediterranean Sea. The two Bays are adjacent to Alexandria City center that is populated with about six million inhabitants, in addition they consider as the two main fishing sources in Egypt. The contaminants are introduced through water way into the Eastern Harbour and El-Mex Bay through several landbased sources. The levels of pollution, particularly heavy metals, have increased dramatically due to the large inputs of land sources (El-Rayis et al. 1997).

At the same time, the Eastern Harbour has been subjected to high levels of pollution due to municipal waste disposal in addition to the waste dumping from ships and shipping activities. Similarly, El-Mex Bay having several industrial plants situated close to the coast and directly discharges its effluents into it. In addition this Bay is an estuarine zone of huge agricultural drain (Omoum Drain), its discharge rate about 2,547.7×10^6 m^3/year (El-Rayis and Abdallah 2006). Few papers have been published concerning heavy metal concentrations in bivalves and fish samples (El-Rayis et al. 1997). On the other hand, numerous studies have been carried out on the heavy metals in sediments and water samples (Abouldahab et al. 1984, 1990, El-Gindy et al. 1986, Dorgham et al. 1987, Rifaat et al. 1996 and El-Rayis et al. 1998).

The objectives of the present study are: (1) to determine the levels of Cd, Cu, Co, Zn, Mn and Fe in algae, mussel, fish and sediment samples collected from different stations of the Eastern Harbour and El-Mex Bay; (2) to compare the present results with the previous ones carried out in the same and different areas; and (3) to study the relation between metal content in two muscle species and in the sediments they inhabit.

Materials and methods

The sampling stations of organisms and sediment in both the Eastern Harbour and El-Mex Bay are shown in Fig. 1. The marine algae species were *Ulva lactuca*, *Enteromorpha compressa* (green algae) and *Jania rubens* (red algae). About 200 g of fresh weight was harvested from the two Bays. The algal samples were cleaned in seawater to remove sand, particulate matter and epiphytes at the laboratory. Finally, samples were rinsed by tap water and distilled water. The samples (50–200 g) were dried at 80°C (to constant weight), homogenized and kept away from metallic materials to avoid contamination. One gram from each algal sample was dissolved in conc. HNO_3 (5 ml) and heated on 70–80°C. Small amount of $HClO_4$ and HNO_3 mixture (1:2) was added very slowly and heated to 120°C. After near dryness, the solution was diluted to 100 ml with 2% HNO_3 in a volumetric flask.

The mussel *Donax trunculus* (31–40 mm shell lengths) and *Paphia textile* (45–55 mm shell length) of 40 individuals was stored in ice insulated box together with fish species taken from fish market in the same day. The box was transferred to the laboratory, then the samples were frozen at −20°C until required for metal analysis. The fish species were Brushtooth lizarfish (*Saurida undosquamis*), Marbled spinefood (*Siganus rivulatus*), Mourmoura (*Lithognathus mormyrus*) and Yellowstripe barracuda (*Sphyraena sphyraena*). The procedure used was that described for digestion of samples in the CEM Digestion Application Manuel (Anon 1994). Fish and mussel tissues samples were digested in microwave oven using HNO_3 acid in Teflon PTFE tube. When the samples became liquid they were diluted to 25 ml with 2% HNO_3 in a volumetric flask.

Sediment samples were collected from four sites at each of the two Bays using Grab Sampler. The collected sediments were composed and sieved, the 0.63 μm size fraction after drying was kept for heavy metal analysis. They were stored in cleaned plastic cups until analysis. The sediment samples were dried at 80°C for 48 h, and dissolved in concentrated HNO_3 acid in Teflon PTFE tubes and small amount of HF acid was added. The subsequent steps of the procedure were followed as for biota samples. The metal concentrations were determined by an Atomic Absorption Spectrophotometer (AAS, Varian Techtron-Model 1250). The concentration values were expressed as mean of three-subsamples, replicates. Blank samples were used inside each batch. The accuracy of the analysis was verified by analyzing the IAEA′s certified reference materials.

Results and discussion

The results of heavy metal concentrations in biota and sediment samples are shown in Table 1. The highest accumulation of metals in macroalgae species at the same sampling site were: Fe, Mn, Zn, Cu and Co in *U. lactuca, E. compressa* and *J. rubens*. Regarding the areas high amounts of Cd, Co, Zn, Mn and Fe were found in *E. compressa* of El-Mex Bay except for Cu in the same species of the E.H.

On the other hand, high amounts of Cd, Cu, Co, Zn and Mn were found in *U. lactuca* of the E.H except for Fe, the highest value was found in *U. lactuca* of EL-Mex Bay. *J. rubens* species was collected only from the E.H, and seems to be in the same range of heavy metal concentrations in both *U. lactuca and E. compressa* except for Co. The E.H. and El-Mex Bay areas had not been investigated before for wide spreading for macroalgae. So when comparing the results reported by Abdallah et al. (2005) in Red Sea on *U. lactuca* with those of the present study we find that Cu, Mn, Co and Zn were increased in *U. lactuca* from the two Bays, but Cd was decreased. Similarly, low levels of Cd, Co, Cu and

Zn were recorded in *U. lactuca* and *Cladophora sp.* from two sites in the Turkish coast of the Black Sea and one site in the southern Baltic Sea (Topcuoglu et al. 2003 and Zbikowski et al. 2007) as compared with the present study in the two Bays. Whereas, Cu, Fe and Mn contents were within the same range in El-Mex and E.H for the same species. In the present study, Cd, Mn, Co and Fe levels in *U. lactuca* from the investigated areas were higher than that in Marmara Sea, while Zn was lower (Topcuoglu et al. 2004).

The two species of bivalves showed different amounts of metals in their tissues (Table 1). The

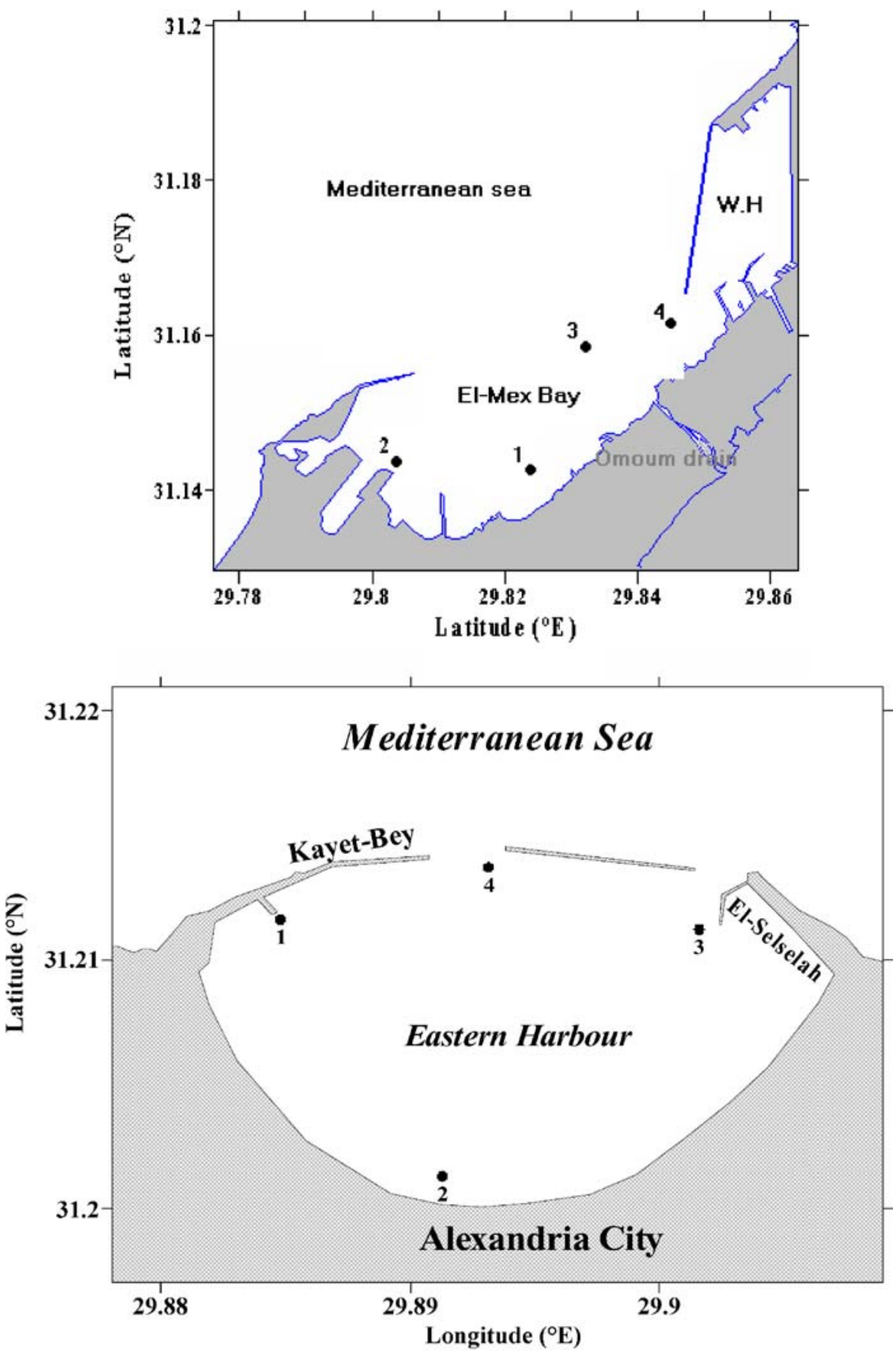

Fig. 1 The sampling stations in El-Mex Bay and the Eastern Harbor

Table 1 Heavy metals concentration (μg g^{-1} dry weight) in biota and sediment samples collected from Eastern Harbour and EL-Mex Bay in 2005

Sample	Cd	Cu	Co	Zn	Mn	Fe
Macroalgae						
E. Compressa[a]	0.77±1.1	20.07±3.2	3.08±1.6	37.78±5.8	134.89±8.7	449.64±36.1
E. Compressa[b]	1.61±1.4	13.77±7.4	15.43±5.5	47.24±3.8	165.65±3.6	1,628.09±44.4
U. lactuca[a]	1.84±0.93	14.52±4.7	7.08±1.3	63.10±3.2	73.95±4.8	514.55±2.1
U. lactuca[b]	0.73±0.24	7.24±2.30	6.58±1.4	27.38±8.3	33.20±3.9	709.23±12.7
J. rubens[a]	0.63±1.2	6.16±1.60	16.60±5.1	28.72±10.4	71.43±11.2	466.65±23.8
Mussel						
d. trunculus[a]	3.93±0.20	12.57±0.22	8.36±0.05	73.99±4.45	21.21±0.23	570.07±8.2
P. textile[a]	4.64±0.28	12.14±0.46	3.07±0.05	100.99±1.50	21.28±0.24	1,052.28±12.4
Fish						
S. undosquamis[a]	1.79±0.36	3.73±0.46	2.93±0.20	15.56±0.74	1.63±0.13	28.19±4.33
L. mormyrus[a]	1.62±0.03	2.42±0.21	1.88±0.15	19.23±0.55	N.D	20.92±1.09
S. sphyraena[b]	1.83±0.05	2.37±0.31	2.59±0.33	18.09±0.41	2.79±0.03	27.83±4.53
S. rivulatus[b]	2.82±0.24	2.75±0.46	4.36±0.94	43.85±2.83	5.96±0.39	55.78±4.18
Sediments						
Sediment[a]	3.80±0.24	14.09±3.24	32.29±4.31	64.49±3.06	95.08±4.63	582.67±15.8
Sediment[b]	8.09±2.8	22.35±1.40	7.87±2.40	305.49±18.9	262.22±10.03	1,787.91±9.34

Sampling site: [a] Eastern Harbour, [b] El-Mex Bay

Dry/wet ratios are 0.14, 0.16, 0.30, 0.26, 0.30 and 0.28 for *D. trunculus, P. textile, S. undosquamis, L. mormyrus, S. sphyraena* and *S. rivulatus*, respectively

pattern of heavy metals occurrence in soft part of the two mussel samples in order of decreasing contents were Fe>Zn>Mn>Cu>Co>Cd for *D. trunculus* and Fe> Zn>Mn>Cu>Cd>Co for *P. textile*. The concentrations of Cd, Zn and Fe were significantly higher ($p<0.05$) in *P. textile* than *D. trunculus*. Whereas, Co level in *D. trunculus* was significantly higher than that in *P. textile*. Because the different species of bivalves have different capacities for accumulation metals (Villar et al. 1999 and Jeng et al. 2000).

Generally, the high concentrations of Cd in the two mussel samples (3.93±0.20 and 4.64±0.28 μg g^{-1}) in

Table 2 Comparison of heavy metal concentrations in fish with values of the other studies (μg g^{-1} dry weight)

Sample area	Cd	Cu	Co	Zn	Mn	Fe
Black Sea coast[a]	<0.02	1.01–4.54	<0.05–0.40	25.7–44.2	0.69–3.56	33–60
Masan Bay–Korea[b]	0.01	0.18–0.25	–	6.33–12.9	–	–
Mediterranean Sea[c]	0.37–0.79	2.19–4.40	–	16.5–37.4	–	19.6–78.4
Iskenderun Bay[d]	0.95	1.57	1.42	4.36	1.71	10.2
Aegean Sea[e]	<0.01–0.04	<0.1	–	<0.5–7.2	–	–
Eastern Harbour[f]	1.7	3.07	2.40	16.29	0.82	24.10
El-Mex Bay[g]	2.32	2.56	3.47	30.97	3.87	41.80

[a] Topcuoglu et al. (2002)

[b] Kwon and Lee (2001)

[c] Canli and Atli (2003)

[d] Türkmen et al. (2005)

[e] Dalman et al. (2006)

[f] Present study

[g] present study

Table 3 Comparison of heavy metal contents (μg g^{-1}) in the surficial sediments of the present and other studies

Sample area	Cd	Cu	Co	Zn	Mn	Fe
Suköy–Marmara Sea[a]	<0.02	12.7	11.1	43.6	372.9	14,896
Atlantic Coast–Spain[b]	0.37	30	–	173.4	–	–
Bay of Güllük[c]	0.56	25	–	81	–	–
Aegean Sea[d]	0.14–0.35	8–28	–	25–72	–	–
Florida Bay[e]	–	15	5	31	614.0	33,000
El-Mex Bay[f]	1.5	44.6	–	60.7	614.0	1,771
El-Mex Bay[g]	8.1	22.3	7.9	305.5	262.2	1,787.9
Eastern Harbour[h]	3.8	14.1	23.3	64.5	95.1	582.7

[a] Topcuoglu et al. (2004)
[b] Usero et al. (2005)
[c] Dalman et al. (2006)
[d] Balci and Küçüksezgin (1994)
[e] Caccia et al. (2003)
[f] El-Rayis et al. (1997)
[g] Present study
[h] Present study

the present study were higher than that in Black Sea and in Atlantic coast of southern Spain (2.88±0.03 and 0.19 μg g^{-1} respectively, Topcuoglu et al. 2004 and Usero et al. 2005) followed by Cu, Co, Mn and Fe. High concentrations of Cd, Cu and Fe were found in mussel samples in the present study as compared with those in *D. trunculus* and *M. corallina* collected from EL-Mex Bay by El-Rayis et al. (1997). Further more, Mn content in mussels from the E.H were lower when compared with the results obtained by El-Rayis et al. (1997).

The results of heavy metal contents in different fish species from the two Bays are also listed in Table 1. The concentration of all metals, except Zn, were significantly higher ($p<0.05$) in *S. undosquamis* than *L. mormyrus* in the E.H. Similarly, the concentrations of all studied metals were higher in *S. rivulatus* than that in *S. sphyraena* in El-Mex Bay. The most abundant metals in the four fish species collected from the investigated areas were Zn and Fe. These levels were also high if compared with those recorded in other fish species studied in the same area (El-Rayis et al. 1997) and in other areas (Türkmen et al. 2005, Canli and Atli 2003 and Kwon and Lee 2001) Table 2. On the other hand, the concentration of Zn, Fe and Cu in fish collected from the Black Sea coast were agreed well with the present results (Topcuoglu et al. 2002). Similarly, the concentrations of Cd, Co, Cu and Mn in the present study were high as compared with those obtained from Black Sea, Masan Bay, Mediterranean Sea and Iskenderun Bay (Table 2). However, these variations of heavy metal

Table 4 Mean concentrations (μg/g dry mass) of metals in sediments and mussels as well as mean bio-sediment accumulation factor values (BSAF) in *D. trunculus* and *P. textile*

	Cd	Co	Cu	Zn	Mn	Fe
Sediment	3.80	14.09	32.30	64.49	95.08	582.67
D. trunculus	4.78	13.43	10.43	92.34	22.67	776.89
P. textile	4.06	10.65	1.95	67.52	18.63	919.89
BSAF-1	1.26	0.95	0.32	1.43	0.24	1.33
BSAF-2	1.07	0.76	0.06	1.05	0.20	1.57

1– for *D. trunclus* 2– for *P. textile*

Table 5 Correlation coefficient between metals in sediments and mussels from the Eastern Harbour

Mussels	Element					
	Cd	Cu	Co	Zn	Mn	Fe
D. trunculus	−0.27	−0.91	0.55	0.26	0.27	0.07
P. textile	−0.24	−0.87	−0.96	1.00	−0.70	0.83

levels in different species depend on the feeding habits, age, size and length of fish and their habitats (Canli and Atli 2003 and Watanabe et al. 2003). The metal representing the lowest mean concentration is Mn in each of *S. undosquamis*, *L. mormyrus* and *S. sphyraena* and Cd in *S. rivulatus* in the two Bays.

The average concentrations of studied heavy metals in sediment are shown in Table 1. The highest amounts of heavy metals in sediment samples were Cd, Cu, Zn, Mn and Fe in El-Mex Bay and only Co in the E.H. When compared the heavy metal concentrations recorded by El-Rayis et al. (1997) at El-Mex Bay with the present study in the same Bay we find that Cd, Zn, Mn and Fe levels seemed increased, but Cu decreased. This is not surprising, considering that El-Mex Bay is the estuary of huge agricultural drain (Omoum Drain), and locating under impact of the highest levels of metal pollution in Egypt (El-Rayis and Abdallah 2006, Abdallah et al. 2007; Abdallah 2007a, b).

Accordingly Cd and Zn (human-related metals) of El-Mex Bay and Co of the E.H showed high concentration in sediments of the present study than those in literature data (Table 3). The concentration levels of Mn and Fe in the sediment of El-Mex and E.H showed averages 262.2±10.03, 95.08±4.63 μg g^{-1} and 1,787.9±9.3, 582.7±15.8 μg g^{-1} respectively. These levels were lower than those measured in sediments of Marmara Sea and Florida Bay. On contrast, Cu contents in the present study were higher than those measured in sediment of Florida Bay and Marmara Sea (Table 3), but lower than the other literature data. The relatively high concentrations of Cd, Cu, Co and Zn in sediments of the two studied Bays can be resulted from anthropogenic influences.

Bioaccumulation of metals in mussels

The efficiency of metal bioaccumulation in two bivalves species was evaluated by calculating the biosediment accumulation factor (BSAF), which is defined as the ratio between the metal concentration in the organism and that in the sediment (Lau et al. 1998 and Szefer et al. 1999). Cd, Co, Zn and Fe sustained the highest mean BSAF for the two studied species (Table 4). Whereas, the lowest BSAF are Cu and Mn. If we compare the two species of studied mussels, we can conclude that *D. trunculus* passes a greater capacity for metal bioaccumulation than *P. textile.* with the exception of Fe. These results was in agreement with Usero et al. (2005).

Relationship between metals in mussels and sediments

Significant correlations ($p<0.05$) for concentrations of Zn and Fe in *P. textile* relative to their concentrations in surface sediments (Table 5). The correlation coefficients of Co, Zn, Mn and Fe were positive but with low levels of confidence ($p>0.05$) in *D. trunculus.* Negative coefficients were found for Cd and Cu in the two species and for Co and Mn in *P. textile,* which indicate that the amount of metals in sediment was not directly reflected in the tissue of *P. textile* and *D. trunculus* except for Zn and Fe of *P. textile.* We can conclude that, the concentrations of some metals in the E.H sediment may probably lower than the concentration that these organisms are able to accumulate in their bodies. For metals of significant correlation ($p<0.05$) the linear regression between the concentration obtained in the bivalves (*Y* value) and those in the sediments (*X* values) were calculated, both expressed as dry weight. The abundance of metals can be ordered as follows in terms of their slope value: Mn (0.48) >Co (0.27) >Fe (0.09) >Zn (0.02) >Cu (−0.16) ≥ Cd (−0.15) for *D. trunculus* and, Zn (1.38) >Fe(0.66) >Co(−0.59) >Cu(−0.78) >Mn (−1.17) >Cd(−1.96) for *P. textile*. The slope was significantly higher than 1 for Zn in *P. textile,* this suggests that the bioavailability of this metal is disproportionately with the degree of Zn enrichment in the sediments (Szefer et al. 1999).

The slopes obtained for *D. trunculus* were greater than those of *P. textile,* except for Fe and Zn, from which we deduced that *P. textile* was less affected by metal pollution than *D. trunculus* in the E.H sediments.

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