

Trace metal distribution in sediments of northern continental shelf of Crete Island, Eastern Mediterranean

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Abstract The present study investigates the distribution of trace metals (Zn, Hg, Cd, Cu, and Pb), as indicators of pollution, in the surficial offshore shelf sediments along the northern coast of Heraklion Prefecture (Crete, Mediterranean Sea). The concentrations and the spatial distribution of the different trace metals, in relation to the sedimentological characteristics and the water circulation pattern of the entire continental shelf, are associated with human inshore sources of pollutants located along the coastline of the study area. Although the trace metal concentrations measured are higher than the background values, they are not considered to be dangerous to human health, as they are lower than the standard values given by the World Health Organisation, with only a few localised exceptions. Furthermore, results reveal the important role of local hydrodynamism that moves fine-grained material and associated trace metals offshore (seawards to wave breaking zone) and then transports them eastwards by entrapping them in the prevailing offshore shelf-water circulation.

Keywords Trace metals · Sediment · Cretan shelf · Pollution · Human health · Aegean Sea

Introduction

Trace metals found in coastal sediment may be derived from a variety of natural and anthropogenic sources (Burrige et al. 1999). In most circumstances, the major part of the metal content in the water and the seabed sediments is anthropogenic and associated with terrestrial sources related to untreated municipal sewage (Shear et al. 1996; Isaac et al. 1997), industrial wastewaters (Majori et al. 1978), intensive aquaculture (Tovar et al. 2000), harbour activities (Tanner et al. 2000) as well as riverine fluxes loaded with urban and agricultural runoff (Pope et al. 1978; Granier et al. 1990).

Relatively high metal contents are associated with finer-grained sediments, mainly clay and silt, which have high absorption capability due to their large specific surface area (Oakley et al. 1981; Cauwet 1987). Furthermore, a strong correlation exists between metals (e.g. Cu, Zn, Fe, Al, Mn, and Ni) and organic carbon content, implying either a primary relationship with grain size (i.e. complexation, adsorption on clay minerals) or a secondary one, as carbon content increases with increasing mud content (Loring 1982; Calvert et al. 1985).

Several studies have been undertaken in the region of Aegean Sea for the determination of trace metals, as indicators of pollution, in coastal sediments. However, most of these studies were undertaken in semi-enclosed river-dominated embayments, such as Evoikos Gulf (Scoullou and Dassenakis 1983; Angelidis and Aloupi 2000), Elefsis Gulf (Scoullou 1983), Pagasitikos Gulf (Voutsinou-Taliadouri and Satsmadjis 1982), the inner part

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of Thermaikos Gulf (Fytianos and Vasilikiotis 1983), Lesvos Island (Aloupi and Angelidis 2001; Gavriil and Angelidis 2005), Gulf of Saros (Sarı and Çağatay 2001), Güllük Bay (Dalman et al. 2006). Similar studies associated with river influxes have also taken place in many other coastal Mediterranean areas, e.g. coasts of Spain (Guieu et al. 1991; Liqueste et al. 2005), coasts of Israel (Herut and Kress 1997; Goldsmith et al. 2001), Gulf of Lions (Morley et al. 1990; Jeftic 1990; Nicolaou et al. 2006).

The present study investigates the distribution of trace metals (Zn, Hg, Cd, Cu, and Pb), as indicators of pollution, in the surficial shelf sediments along the northern coast of Heraklion Prefecture (Crete), an area characterized by an exceptionally well-developed tourist infrastructure not only for Greece but also for the entire Eastern Mediterranean. Moreover, trace metal spatial concentration is discussed in relation with the prevailing sedimentological characteristics, the shelf water circulation pattern and the different potential terrestrial sources of pollutants, i.e. influxes of several torrential rivers, the urban sewage outfalls of Heraklion city and its industrial area and finally the Heraklion harbour. As the present study is based on a data set collected in early 1990s (1991), it could be used for a comparison of current and future changes in the environmental status of this particular coastal area. Such changes could be or may have been induced by the increased tourist activities, the operation of biological treatment plant of Heraklion metropolitan area and the operation of the outfall of the major Electric Power Station of Crete.

The study area

Physical geography

The study area is part of the northern continental shelf of Crete island, extending to depths between 10 and 70 m

along the coastline of the Prefecture of Heraklion and lying between the Cape Panagia, to the west, and the Malia Bay, to the east (Fig. 1). Geologically, it is located on the southern continental margin of the central part of Cretan Sea. The shelf area of the northern Cretan continental margin is very narrow (<10 km) and relatively steep (slope gradient of 1.5°), becoming wider eastwards. The shelf break occurs generally at a water depth of between 100 and 150 m, followed by a relatively steep slope with gradients of between 2° and 4° and with variable morphology (Chronis et al. 2000).

As far as surface sedimentology of the study area is concerned, there is little information for the nearshore zone (water depths <12 m) (Poulos et al. 2002) as well as the continental shelf and the upper slope (>10 m) of Heraklion Bay (Dounas and Papadopoulou 1993; Chronis et al. 2000). The nearshore zone is characterised by sandy material due to grain-size sorting which removes fine fractions seawards while the outer part of the continental shelf is dominated by muddy sediments (silt and clay), mainly of terrigenous origin. Along the coastline a hinterland area of 1,670 km², including some low-lying Quaternary coastal plains and the catchments of five small rivers, is drained (Table 1).

Oceanography

Waves and tides

The northern Cretan continental margin is an essentially tideless environment, with an average tidal range <10 cm (Tsimplis 1994). However, meteorologically induced sea level rise, usually associated with strong north winds, can exceed 0.5 m (Ghionis et al. 2004), reaching a maximum of 1 m (HNHS 2005).

The study area is mainly influenced by wind-generated waves of N, NE and NW origin, with the NW winds prevailing both on monthly and annual base (annual frequency

Fig. 1 Bathymetry (in metres) along the northern coast of Heraklion Prefecture (Crete, Eastern Mediterranean). Locations of river mouths (a Almiros, b Gazanos, c Xiropotamos, d Giofiros, e Karteros, f Aposselemis) and of the major human establishments (black boxes) regarded as possible sources of pollutants (1 oil storages, 2 Electric Power Station; 3 and 4 waste treatment sewers, 5 former military base of Gournes)

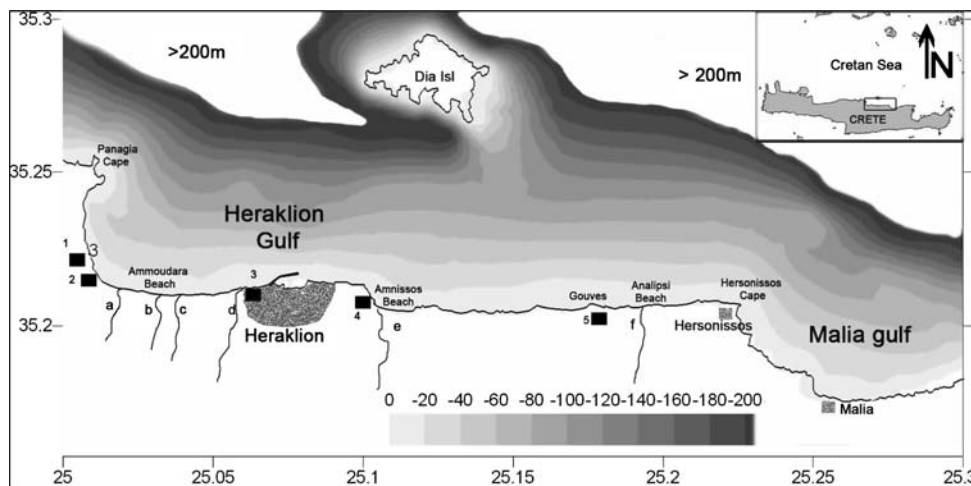


Table 1 Drainage area, maximum elevation and mean annual water discharge of the fluvial systems discharging along the coastline of the study area

Fluvial systems	Area (km ²)	Water discharge (10 ⁶ m ³ /year)	Maximum elevation (m)
Gazanos	192.0	15.6	380
Xiropotamos	35.0	–	280
Giofiros	279.0	21.6	428
Karteros	208.3	–	–
Aposselemis	120.5	–	1,300
Other streams	835.8	–	<50
Total	1670.4		

of 28.89%), except for the winter period (November to January) when waves approach primarily from the North.

The highest predicted wind-generated waves, originating from the North, reach heights of up to 6 m with corresponding periods of 11 s. The most frequent incoming waves, from NW and NE, usually reach a height of usually <1 m and last for periods of <5 s (Ghionis et al. 2004). On the basis of the highest incoming waves, during storms, the closure depth (which denotes the limit of the shore zone with respect to the mobility of the seabed sediment), lies at depths of 11.6, 4.7 and 3.5 m for the waves approaching from the N, NE and NW directions, respectively. Therefore, the offshore zone is considered as starting at water depths >12 m.

Shelf water circulation

The northern coast of Crete is influenced by the variable circulation of Cretan Sea which consists of a series of gyres, eddies and small-scale structures, interconnected with currents and jets (Theocharis et al. 1999). Some of these features are permanent and others seem to be transitional or recurrent. The complex circulation pattern along with the prevailing northerly and north-westerly winds, determines the surface water mass transport of the inner shelf.

A synthesis of all existing information on the Cretan Sea and its shelf circulation pattern (Theocharis et al. 1993; Dounas and Papadopoulou 1993; Karakassis 1994; Drakopoulos 1996; Kornilios et al. 1998; Theocharis et al. 1999; Georgopoulos et al. 2000; Nittis et al. 2002; Cardin et al. 2003; Triantafyllou et al. 2003; Hamad et al. 2006) is summarized in Fig. 2. Although the circulation pattern of the study area is highly variable, some permanent features can be identified. A permanent cyclonic gyre (Cretan cyclonic gyre) is established at the eastern part of the Cretan Sea. During high stratification conditions of the water column (observed during the summer period and most of the winter periods after 1994, Zervakis et al. 1998), an anticyclone appears in

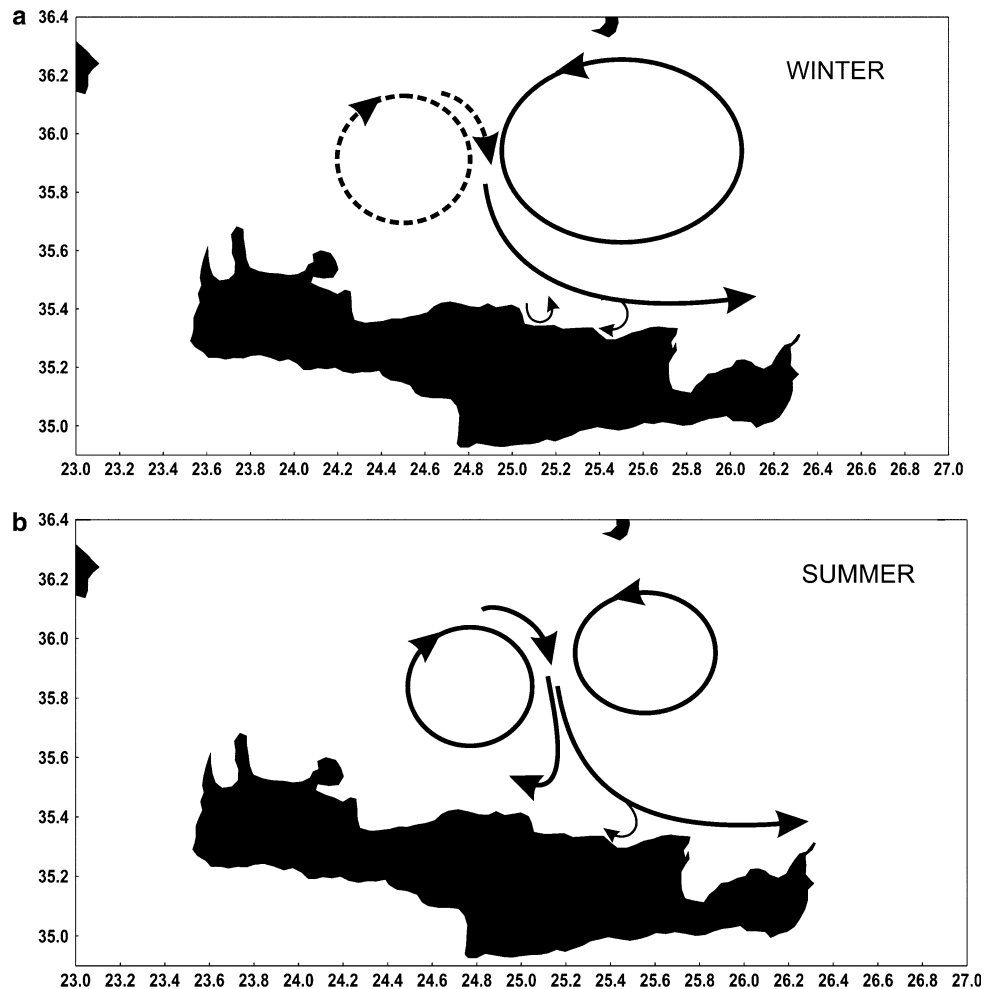
the central-western part of the Cretan Sea (West Cretan anticyclone) and, with the Cretan cyclonic gyre, forms a mesoscale dipole. The presence of this dipole results in a south, southeastern flow towards the shelf. In winter, however, the cyclonic gyre usually appears enlarged and extends to the central Cretan Sea. Thus, during periods of low anticyclonicity, the flow over the shelf is E-SE becoming more energetic when the anticyclone is present.

According to the above-mentioned observations, the circulation along the north coast adjacent to Heraklion city is mainly dictated by the general circulation pattern of the Cretan Sea and the local bathymetry and to a lesser extent by the prevailing wind conditions. Thus, during winter, the S-SE flow over the shelf as it approaches to the coast is topographically stirred to a clear eastward flow along isobaths. On the local scale a cyclonic eddy has been reported within the western part of Heraklion Bay and an anticyclone in Malia Bay (Dounas and Papadopoulou 1993; Karakassis 1994). In summer, depending on the position of the dipole, the southward flow bifurcates near the coast and can be both eastward and westward (Dounas and Papadopoulou 1993). Finally, current metre records in the study area showed that local currents are not very strong (~10 cm/s) and 97% of the variability is along isobaths (Nittis et al. 2002). On an annual basis, the net transport is weak and towards the east, whilst spectral analysis indicates that processes with periods of less than two days (e.g. inertial currents, tidal currents, etc.) are energetically insignificant with no effect on the local water circulation.

Human activities: potential sources of pollution

The coastal region under investigation, including Heraklion city which is the administrative capital of Crete, is the most important tourism centre of Crete. This area is also highly industrialised. Among the various human infrastructures and activities that influence coastal waters and associated seabed sediment, the most important are the following: (1) The Electric Power Station, which utilises sea water for cooling, while tankers anchored at its nearshore area supply crude oil; (2) the biological treatment plant of Heraklion metropolitan area which during the sampling period discharged waste waters in Giofiros river (1 km upstream of its mouth); (3) the port of Heraklion, which is the third, as far as commercial importance is concerned, in Greece and one of the largest in the eastern Mediterranean; (4) the mouths of five small rivers and a number of smaller streams, which drain, in total, a hinterland area of about 1,700 km², carrying urban wastes (originating from the various inland villages), fertilisers and agrochemicals produced by agricultural production, and wastes from cattle-raising units; (5) the international airport of Heraklion, constructed very close to the coastline and with its

Fig. 2 Schematic presentation of the upper thermohaline circulation in Cretan Sea, during the winter (a) and summer (b) periods (dashed lines refer to recurrent features)



main runway extending over Amnisos Bay; and (6) the different tourist establishments built along and/or very close to the coastline, being responsible for the pollution of the nearshore waters (Fig. 1).

Data collection and analytical methods

Sampling procedure

A field survey was carried out at 88 stations over 21 transects from 10 to 70 m depths perpendicular to the coastline of Heraklion and Malia Bays (Fig. 3). Sampling was carried out in September 1991 using the R/V *Philia*. The positions of each station were calculated using a FURUNO SFN-70 satellite navigator and depth was measured by the SIMRAD K-400 echo sounder. At each station, a single undisturbed sediment sample was taken using a 0.1 m² top opening Smith McIntyre grab (Smith and McIntyre 1954). Redox potential measurements were taken at 1-cm increments using a conventional combined electrode. From each sediment sample, three sub-samples of

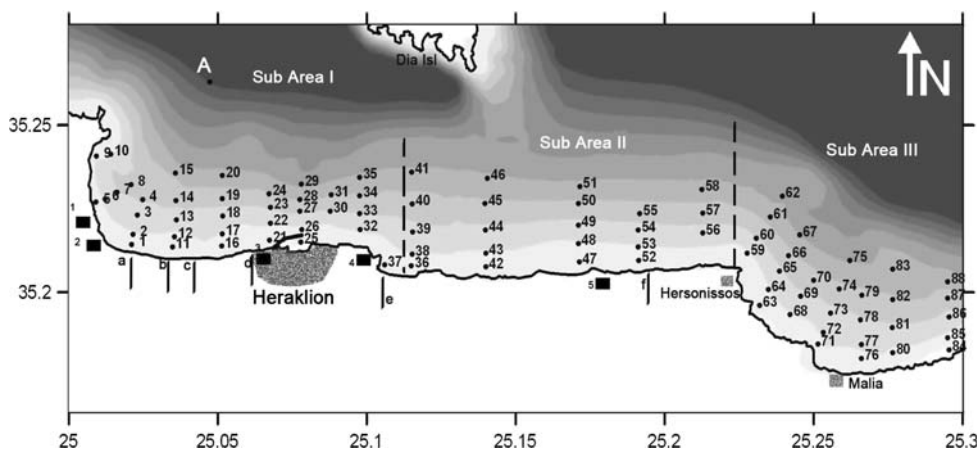
surface sediment (0–2 cm) were collected by means of plastic corers (2.2 cm diameter) for trace metals, chloroplastic pigment equivalent and particulate organic carbon analysis as well as grain-size analysis. The sediment sub-samples were frozen and stored on board at –20°C until analysis upon return to the laboratory.

Laboratory analyses

In the laboratory the sediment samples were oven-dried at 60°C and after homogenization, representative portions of the samples were separated for grain size analyses. The standard sieve analysis technique (Folk 1980) was used to determine the sand and mud (silt and clay) percentages, of each of the surficial sediment samples. Chloroplastic pigment equivalent (CPE) and particulate organic carbon (POC) in sediment samples were determined according to the techniques outlined by Strickland and Parsons (1972) with the modifications described by Basford and Eleftheriou (1988).

For the determination of the heavy metals, sediment samples were separated using a 63- μ m plastic sieve, into

Fig. 3 Locations of (i) sampling stations (1–88), (ii) offshore reference station A; (iii) river mouths (a–f) and (iv) major coastal human activities (1–5 black boxes) along the northern coast of Heraklion Prefecture



two classes: sand (>63 μm) and mud (<63 μm). This approach permitted the quantitative analysis of metals in both sand and mud (silt and clay) fractions and a satisfactory degree of normalisation of the results, in order to compare trace metal concentrations from samples of different granulometry. The total concentrations of heavy metals (copper, zinc, mercury, lead and cadmium) in the surface sediments were determined by means of Atomic Absorption Spectrophotometry (AAS) with background correction. The Graphite Furnace AAS was used in the determination of copper, lead, cadmium and zinc levels and the Cold Vapor AAS for the determination of mercury in the prepared samples. The sediment samples were prepared by freeze-drying, digestion with HNO₃ 65%: H₂O₂, centrifugation and dilution with nanopure water. Together with the samples, certified reference material from the Canadian National Bureau of Standards (BCSS-1 and PACS-1) was analysed in order to control the method used. During the determination of heavy metals with AAS a matrix modifier (NH₄H₂PO₄ 5%) was used. All the material used during the analysis was precleaned with HNO₃.

Statistical analyses

In order to examine to which extent the sediments are polluted, the enrichment factor (Ef) has been calculated (concentration of the metal/standard value of the metal for the Mediterranean Sea according to World Health organization, WHO). Principal component analysis (PCA) was performed, using Euclidean distances, in order to identify any meaningful patterns and associations of the trace metals and the most important environmental factors measured across samples. Data were normalised prior to analysis in order to have comparable (dimensionless) scales. All variables were included at the same time in the analysis since there was no high correlation between the variables ($\rho \geq 0.9$) according to Spearman’s correlation coefficient. Spearman’s coefficient was also applied in

order to explore any correlation between the PCA axes scores and the values of the environmental data. These relationships can largely contribute to the verification of any significant pattern derived from PCA analysis. The PRIMER v6 and STATISTICA statistical software packages were used for the above-mentioned data analyses.

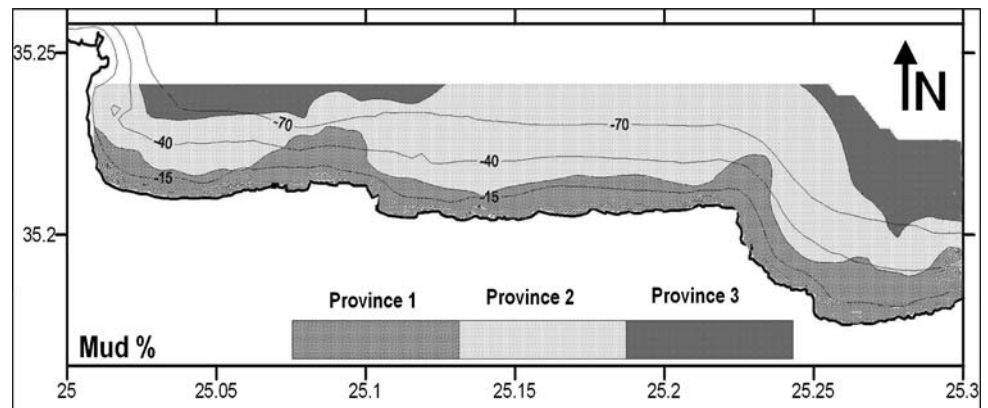
Results and discussion

Sediment texture and redox potential

The grain-size distribution and relevant textural classification of the surficial sediments of the continental shelf along the coast of the study area is presented in Fig. 4 and also tabulated in Appendix I. In relation to their sand and mud (silt and clay) contents, the shelf surficial bottom sediments can be divided into three sedimentary provinces: Province 1 (water depths <15 m), where the bed sediments were sandy (sand content >90%); this zone more or less coincides with the zone being under the influence of the incoming waves, except for the offshore areas seaward to the mouth of River Giofiros and Heraklion port as well as the promontory of Cape Hersonissos, where the sandy bed reached 30 m of water depth. Province 2, lying between depths of 20 and 40 m, has the characteristics of a transitional zone between the nearshore sandy zone and the outer muddy shelf. Province 3 (outer shelf), extending to water depths >40 m, consisted of mud (silt and clay >90%); this dominance of fine-grained material is related to the offshore distribution of river plumes, wave-induced re-working and subsequent offshore dispersion as well as the advection of fine material by the inner shelf water circulation pattern.

Redox potential (Eh) showed positive values ranging from 39 to 601 mV for the top 2 cm of sediment in the study area (Appendix I). Surface sediment redox potential values generally decreased with depth presenting high values in the shallower part of the continental shelf of the

Fig. 4 The three sedimentary provinces (1–3) according to the spatial distribution of mud (silt + clay) percentages



study area (10 m), due to the strong hydrodynamism that characterizes this zone, and lower values in the deeper part (20–70 m). Most of the lowest values (<100 mV) were observed over the western part of the area.

Particulate organic carbon and chloroplastic pigment equivalent

Particulate organic carbon content in surface sediments of the study area varied between 0.02 and 10.13 mg/g, with the majority of sediment samples presenting concentrations <5 mg/g (Fig. 5a, Appendix I). The increasing POC content seawards indicates a strong association with fine-grained sediments, while the overall low concentrations throughout the study area denote the terrigenous origin of the shelf sediment. Increased POC contents (>5 mg/g) were observed over the western part of the study area and, in some cases, over its central and eastern parts. These relatively high concentrations may be related to local terrigenous inputs (e.g. river fluxes) and the overall water circulation pattern. The latter may be the cause of some relatively high POC concentrations measured in Malia Bay, favoured by the presence of the anticyclonic gyre (Fig. 2).

Chloroplastic pigment equivalent (CPE) in the surface sediments of the study area ranged from 0.7 to 35 $\mu\text{g/g}$ with most of the samples presenting concentrations <10 $\mu\text{g/g}$ (Fig. 5b, Appendix I). The overall low concentrations of CPE were observed in the shallower part of the continental shelf of the study area (10 m). The highest CPE values were observed in the deeper zone of the study area and especially over its western part.

Trace metals

Spatial distribution

Trace metal concentrations for all the surficial sediment samples are tabulated in Appendix I and their minimum, maximum as well as weighted-average values are given in

Table 2. In general, copper, lead and zinc showed the highest concentrations at water depths of 20–40 m (Fig. 6).

In particular, copper showed the highest concentrations in the western part of the study area (46–126 ppm) and in water depths of 20–40 m (Fig. 6). These high values can be attributed to the outfall of the Electric Power Station, which is located at the backshore zone, and to cargo ships carrying crude oil (Fig. 1). Over the central and eastern parts of the study area, the Cu values were lower than 20 ppm, except for some locations presenting values of up to 37 ppm most probably due to some localised terrestrial inputs (i.e. anthropogenic influence) and to water circulation, as in the case of the Malia Bay (depths of 30–40 m).

The highest concentrations of lead (40–60 ppm), zinc (60–125 ppm) and cadmium (0.9–1.9 ppm) were measured offshore of the zone extending along the coast of Heraklion city and port (Fig. 6). Low values of these trace metals (Pb 8–25 ppm, Zn 7–31 ppm, Cd 0.04–0.25 ppm) were observed at the distal coastal areas to the west and the east of Heraklion city, indicating areas of low human impact. Cadmium also showed increased values in the area surrounding the harbour of Heraklion city (1.22–1.86 ppm), being attributed to intensive maritime activities (10 m depth).

Mercury showed high concentrations (0.5–2.0 ppm) at the western end of the central part of the study area (Fig. 6), and particularly in front of the outfall of the waste treatment plant of the industrial zone of Heraklion city, which is located at a water depth of about 10 m. The distribution pattern showed a second area of increased Hg concentration (1.16 ppm) in front of the former military base of Gournes in water depths of 10 m (Fig. 1) most probably due to military wastes disposed locally.

According to the spatial distribution of trace metals concentrations, the study area can be divided into three sub-areas: the western (I), the central (II) and the eastern (III) (Fig. 3):

Sub-area (I) is located over the western part of the area under investigation, extending from Cape Panagia to Amnissos beach, where most of the main sources of potential

Fig. 5 Distribution of (a) particulate organic carbon (POC in mg/g) and of (b) chloroplasic pigment equivalent (CPE in µg/g) in the surficial shelf sediment of the study area

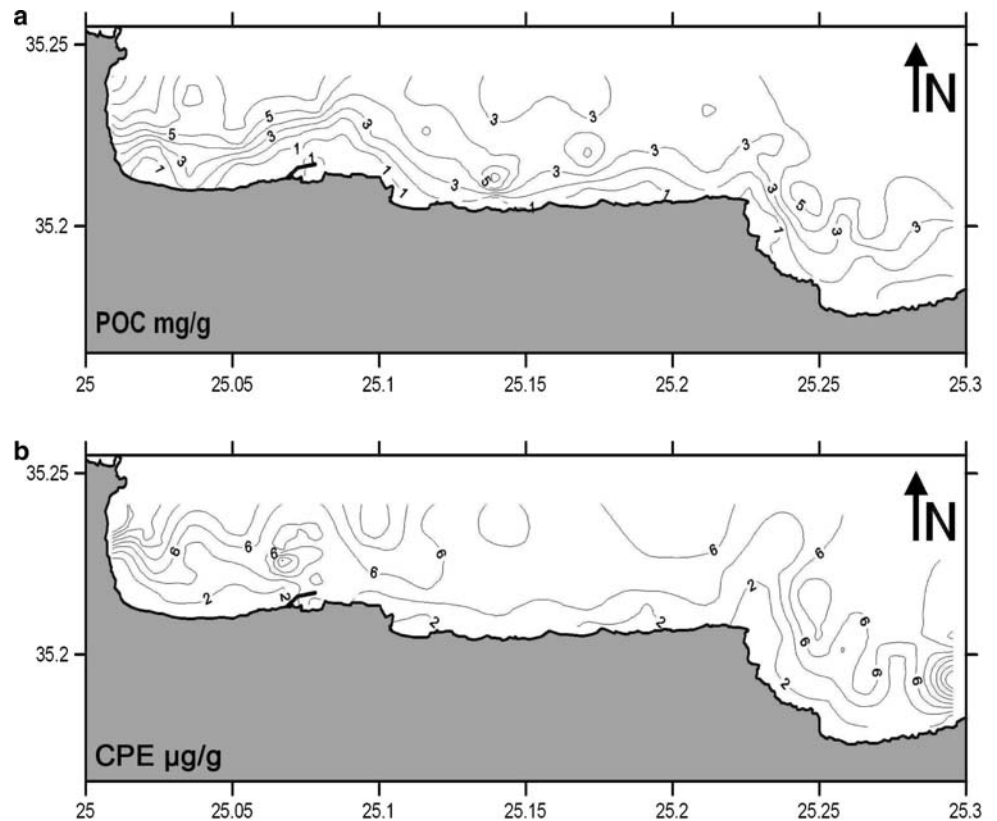


Table 2 Minimum, maximum and averaged values of trace metal concentrations (in ppm) in the three sub-areas and the study area as a whole

	Sub-area I			Sub-area II			Sub area III			Total study area		
	Min.	Max.	Aver.	Min.	Max.	Aver.	Min.	Max.	Aver.	Min.	Max.	Aver.
Cu	13.65	126.10	46.20	3.01	37.30	21.68	2.07	29.5	8.39	2.07	126.1	27.11
Hg	0.03	0.21	0.06	0.04	2.04	0.50	0.03	0.25	0.11	0.03	2.04	0.19
Pb	10.40	63.00	34.99	10.31	33.26	20.44	8.26	25.79	14.33	8.26	63.00	24.26
Cd	0.04	1.86	0.44	0.04	0.97	0.25	0.04	0.10	0.07	0.04	1.86	0.27
Zn	29.63	125.32	72.24	7.99	50.01	31.66	6.89	32.4	15.93	6.89	125.32	42.74

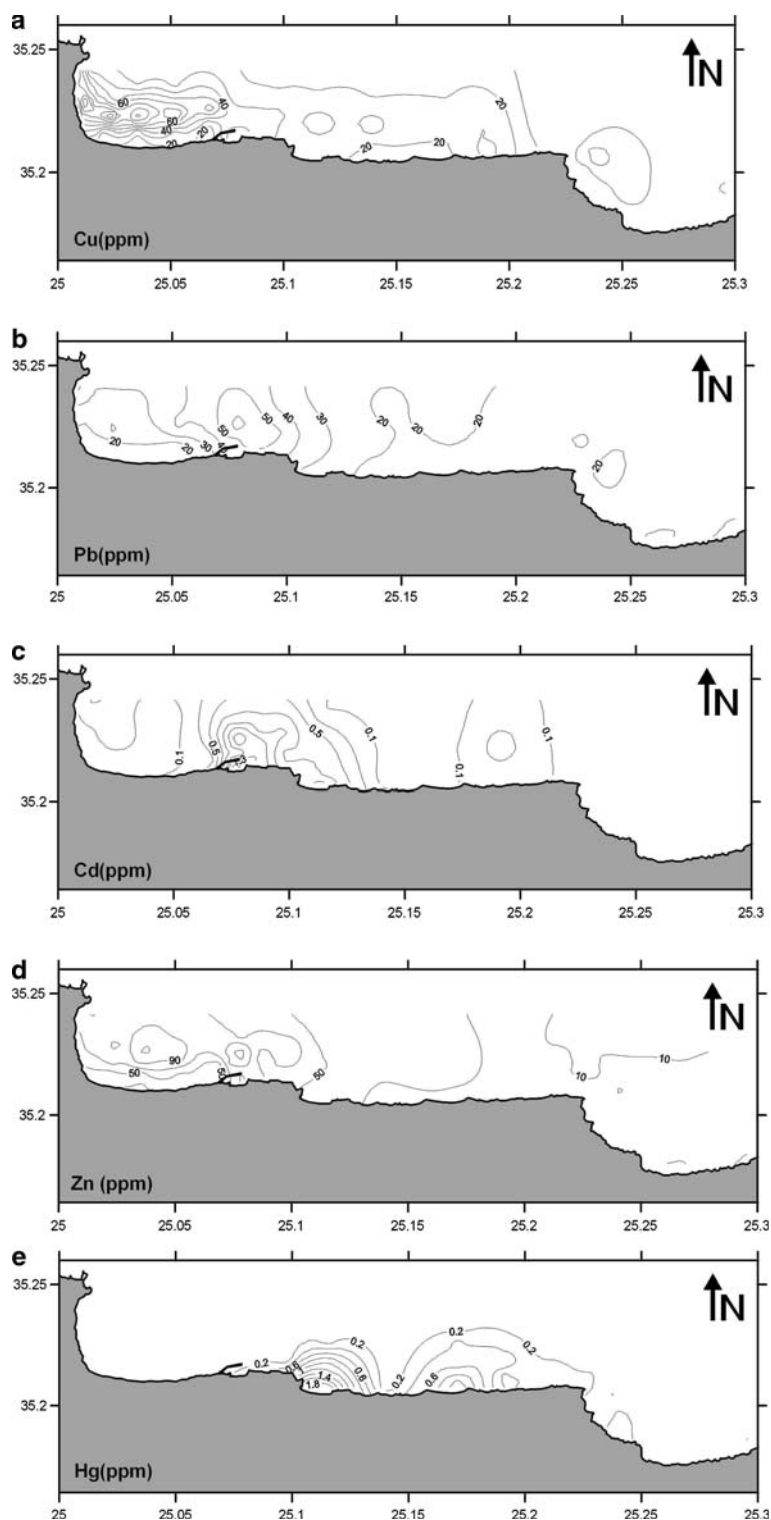
pollution are present, i.e. the sewage outfall of Heraklion city, petrol storing facilities, the outfall of the Electricity Power Station, the harbour zone and the urban waste sewers. Over this section all of the metals examined, except for Hg, presented the highest values and especially at water depths of 20–40 m (Table 2). This sub-area can be divided further into (a) the western part, where the main source of trace metals is the Electricity Power Station and Cu presented the highest concentrations in water depths of 20–40 m, decreasing eastwards and (b) the eastern part, where pollutants were associated almost exclusively with the City itself and the harbour (Pb, Zn and Cd presented the highest values in this part).

Sub-area (II), extending from Ammissos beach to Hersonissos Cape (Fig. 3), where the coastal zone is characterised generally by lower values of trace metals in comparison with sub-area I (Table 2). An exception to this

general pattern is the distribution of Hg concentrations, which presented here the highest values in two sites (1.16 and 1.92 ppm, respectively): in front of the former military base of Gournes area and adjacent to the outfall of Heraklion industrial area. The latter being very close to the coastline and at a water depth of 10 m (Fig. 1) appears to pollute locally, without affecting the rest part and eastward-lying part of the sub-area II.

Sub-area III lies to the east of Cape Hersonissos and Malia Bay (Fig. 3), which is associated with an anticyclonic circulation of surface waters. Though trace metals concentrations generally showed the lowest values, there are no great human infrastructures except for different tourist establishments built along and/or very close to the coastline in this sub-area; this is a consequence of the eastward-moving waters (in sub-area II) and their subsequent entrapment by the anticyclonic gyre of Malia Bay.

Fig. 6 Trace metal concentrations (in ppm) in the surficial shelf sediment of the study area (**a** copper, **b** lead, **c** cadmium, **d** zinc and **e** mercury)



Enrichment factor (E_f)

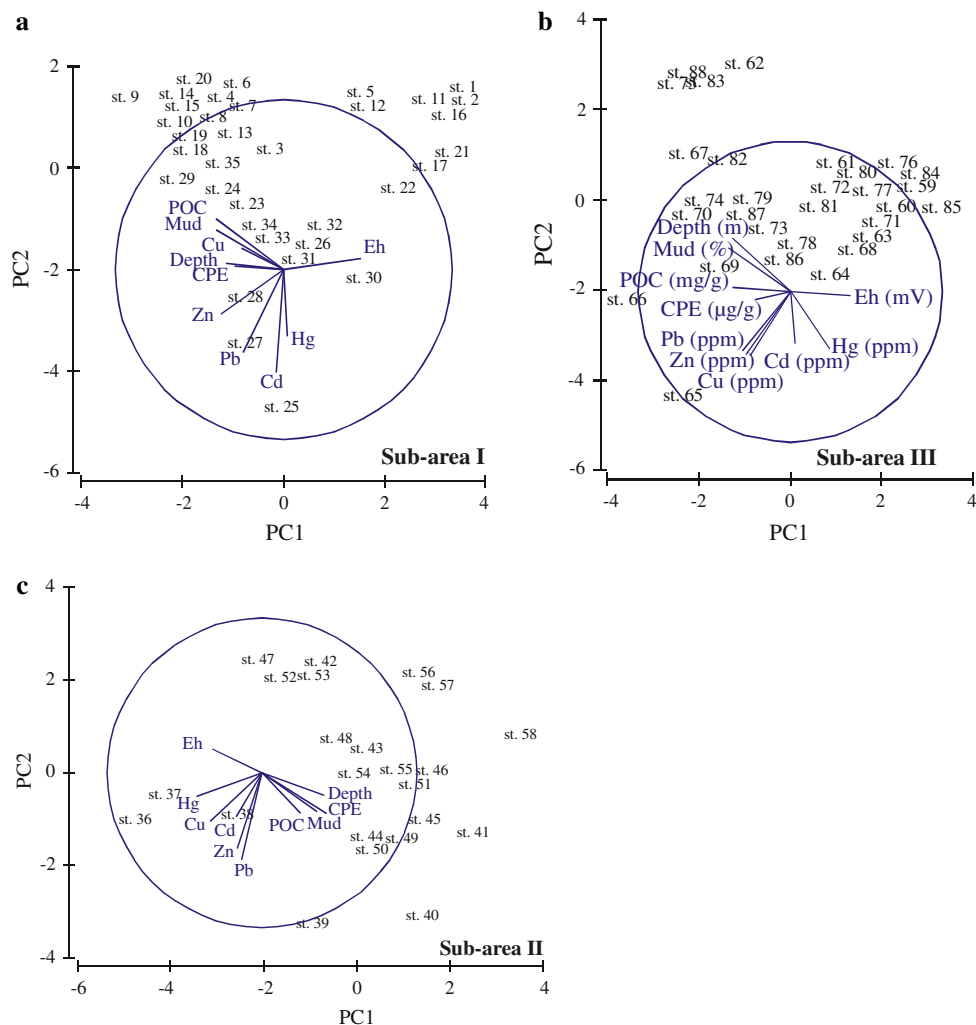
In order to examine the extent to which the shelf surficial sediment of the central part of Cretan shelf is polluted, a comparison was made between the measured concentrations and the standard values presented for Mediterranean Sea by

the World Health Organisation (WHO 1995) as well as the values obtained from reference station A of the Cretan continental slope (water depth of 200 m) by calculating the enrichment factor for each sub-area (Table 3). Reference station A is considered here as unaffected by human activities (“clean”) and a geochemically representative station

Table 3 Minimum, maximum and averaged values of enrichment factor of trace metal concentrations in each sub-area (S-A I, II and III) with respect to standard values of the WHO (1995) and values calculated from the reference station (A) of the Cretan continental margin

Metal	WHO (1995)					Reference station A					
	Cu	Pb	Cd	Zn	Hg	Cu	Pb	Cd	Zn	Hg	
Standard values	13	38	1	45	0.3	16.77	0.23	0.13	48.49	0.03	
S-A I	Min.	1.05	0.27	0.04	0.66	0.11	0.81	1.30	0.27	0.61	1.10
	Max.	9.70	1.66	1.86	2.78	0.69	7.52	7.88	14.31	2.58	6.93
	Aver.	3.55	0.92	0.44	1.61	0.21	2.75	4.37	3.38	1.49	2.09
S-A II	Min.	0.23	0.27	0.04	0.18	0.12	0.18	1.29	0.28	0.16	1.17
	Max.	2.87	0.88	0.97	1.11	6.79	2.22	4.16	7.44	1.03	67.9
	Aver.	1.67	0.54	0.25	0.70	1.65	1.29	2.56	1.90	0.65	16.53
S-A III	Min.	0.16	0.22	0.00	0.15	0.11	0.12	1.03	0.00	0.14	1.13
	Max.	2.27	0.68	0.1	0.72	0.82	1.76	3.22	0.75	0.67	8.20
	Aver.	0.65	0.38	0.06	0.35	0.37	0.50	1.79	0.48	0.33	3.71

Fig. 7 Principal component analysis for abiotic data [water depth, trace metals, mud content (%), redox potential (*Eh*), particulate organic carbon (*POC*) and chloroplactic pigment equivalent (*CPE*)] of sub-area I (a), sub-area II (b) and sub-area III (c)



for Cretan continental margin, where the study area is included.

It is worth noticing the differences between the standard values presented by WHO (1995) and those obtained from

Reference Station A (Table 3). The latter are characterised by much lower values of Pb, Hg and Cd, by up to one order of magnitude, than the standard values, while concentrations of Cu and Zn were slightly higher (but less than 10%).

Table 4 Eigenvalues and % variation of PCA of the three sub-areas (I, II and III)

PC	Eigen-values	Variation (%)	Cumulative % variation
Sub-area I			
1	3.68	36.8	36.8
2	2.43	24.3	61.2
3	1.46	14.6	75.8
4	0.88	8.8	84.5
5	0.68	6.8	91.3
Sub-area II			
1	4.03	40.3	40.3
2	2.69	26.9	67.2
3	1.08	10.8	78.0
4	0.78	7.8	85.8
5	0.59	5.9	91.7
Sub-area III			
1	3.85	38.5	38.5
2	2.46	24.6	63.1
3	1.11	11.1	74.2
4	0.82	8.2	82.4
5	0.60	6.0	88.4

Taking into account WHO values, sub-area I may be regarded as affected by pollution, with most of the Ef values being above unity (>1) with the exception of Hg, presenting 0.69 as maximum and 0.21 as averaged Ef values. Sub-area II presented Ef factors >1 only for Cu (max. 2.87, aver. 1.67) and Hg (max. 6.79, aver. 1.65), while sub-area III appears to be the 'cleanest' region, characterised by the lowest values of enrichment factor, except for the maximum value of Cu (max. 2.27).

With respect to the values obtained from the Reference Station A, most of the Ef values, were >1, especially for the first and second sub-areas, i.e. the western and central parts of the study area. The fact that all the shelf sediment presented high concentrations of trace metals, in comparison with the ones obtained from Station A (upper continental slope), indicates the presence of pollutants in the study area, but within the safe limits according to human health established by WHO (1995). Exceptions to this general situation are a few 'hot spots' located in the inshore zone.

Principal component analysis

In order to investigate further the relationship between the observed trace metal concentrations and the prevailing environmental conditions (e.g. bathymetry, sedimentological characteristics) as well as the potential anthropogenic sources of pollution, PCA for abiotic data of each sub-area was applied separately (Fig. 7).

As far as sub-area I is concerned PCA showed that the first principal component axis was characterized by the opposition between redox potential and mud percentage and organic carbon along with depth, accounted for almost 37% of the variability (Tables 4, 5). The second axis, characterized by Pb and Cd concentrations, accounted for an additional 24% of the variability and the third principal component axis, characterized by Cu concentrations, accounted for an additional 15% of the variability (Tables 4, 5). Results for sub-area I showed that the deeper muddy stations (30–70 m) grouped in the upper left part of the PCA graph were mainly associated with increased POC and Cu concentrations. The highest concentrations of Pb were particularly present in the stations of intermediate depth (20–40 m) and lower concentrations in the deeper muddy ones. Additionally, increased Cd concentrations were present in the stations of intermediate depth of Heraklion port (positioned in the lower part of the graph) probably due to intensive maritime activities. Shallow sandy stations (10–20 m) characterized by the highest values of redox potential and present in the upper right part of the graph were not really associated with high trace metal concentrations.

For sub-area II, the first two axes of PCA explained 67% of the total variance (Tables 4, 5). The first axis accounted for 40% of the total variance, opposed depth and mud percentage on one side and Hg concentrations on the other, while the second axis was characterized by Pb and Zn concentrations (27% of the variability). In this sub-area PCA analysis showed that increased Hg concentrations were present in the shallow sandy stations in front of the former military base of Gournes area and adjacent to the outfall of Heraklion industrial area positioned in the left part of the graph. On the other hand, increased Pb and Zn concentrations were generally associated with muddy deeper stations and also with the sandy and muddy stations of the western part of the area (positioned in the lower part of the graph).

Finally, for sub-area III the first two axes explained 63% of the total variance (Tables 4, 5). The first axis was characterized by the opposition between redox potential and mud percentage and particulate organic carbon along with depth (38% of the variability). The second axis was characterized by Cu and Zn concentrations (25% of the variability). Results derived from PCA analysis of this sub-area revealed that muddy stations are associated with high POC concentrations and positioned in the left part of the graph, while the highest Cu and Zn concentrations were associated with stations of intermediate depth (20–40 m) and particularly of muddy sand bottoms (positioned in the lower part of the graph).

Spearman's coefficient verifies the different patterns derived from PCA analysis for the three sub-areas (Table 6).

Table 5 Eigenvectors of PCA of the three sub-areas (I, II and III)

Eigenvectors Variables	Sub-area I					Sub-area II					Sub-area III				
	PC1	PC2	PC3	PC4	PC5	PC1	PC2	PC3	PC4	PC5	PC1	PC2	PC3	PC4	PC5
Depth (m)	-0.346	0.035	-0.544	0.148	0.135	0.403	-0.148	0.334	0.108	-0.387	-0.377	0.354	0.098	0.185	-0.158
Eh (mV)	0.460	0.067	-0.130	-0.116	0.069	-0.322	0.152	-0.271	-0.485	-0.522	0.395	-0.029	0.231	0.268	-0.308
Mud (%)	-0.402	0.231	-0.272	0.180	0.355	0.418	-0.264	0.182	0.014	-0.175	-0.405	0.292	-0.102	0.181	0.218
CPE (µg/g)	-0.295	0.026	0.131	-0.828	-0.006	0.367	-0.234	-0.122	-0.215	0.396	-0.233	-0.048	0.699	-0.339	-0.415
POC (mg/g)	-0.399	0.299	0.087	-0.207	0.155	0.247	-0.264	-0.417	-0.590	0.105	-0.383	0.026	-0.085	0.610	-0.282
Cu (ppm)	-0.245	0.133	0.629	0.198	-0.194	-0.170	-0.288	-0.591	0.526	0.140	-0.282	-0.427	-0.248	-0.138	-0.128
Hg (ppm)	0.025	-0.391	0.339	0.043	0.815	-0.423	-0.152	0.331	-0.180	0.105	0.260	-0.385	-0.255	0.311	-0.343
Pb (ppm)	-0.238	-0.492	-0.182	-0.124	-0.274	-0.129	-0.564	0.111	-0.072	-0.095	-0.307	-0.396	0.038	0.005	-0.252
Cd (ppm)	-0.046	-0.609	-0.111	-0.114	-0.023	-0.335	-0.313	0.335	-0.146	0.377	0.028	-0.348	0.543	0.469	0.540
Zn (ppm)	-0.375	-0.261	0.180	0.367	-0.223	-0.162	-0.489	-0.099	0.154	-0.445	-0.307	-0.420	-0.089	-0.197	0.303

Eh redox potential, *mud* = silt + clay, *CPE* chloroplastic pigment equivalent, *POC* particulate organic carbon

The general pattern derived from PCA for all the three sub-areas revealed that the finer-grained sediments in the deeper zone of the study area were associated with increased particulate organic carbon, while trace metal distribution seem to be associated partly with granulometry and partly with a few localised anthropogenic sources of pollutants.

Conclusions

The coastal area between Heraklion and Malia Bays on the basis of trace metals spatial distribution and the position of the terrestrial sources of pollution is divided into three distinct sub-areas: the western sub-area where most of the main sources of pollution are located, the central sub-area and the eastern sub-area. In the western sub-area, increased Cu concentration is mainly associated with the operation of the largest Electric Power Station of Crete and Pb–Zn–Cd–Hg concentrations are induced by the sewage of Heraklion City and the industrial zone. In the central sub-area trace metal concentrations were lower in comparison with the western sub-area, except for the increased Hg concentrations found at the dumping site of wastes in front of the former military base at Gournes. Although the eastern sub-area presents the lowest values of trace metal concentrations of the study area, it would be expected to present even lower values because there are no important sources of pollution there. Trace metal concentrations in this area are probably found due to the entrapment by the Malia anti-cyclone gyre of eastward moving heavy metals.

The observed pattern of the environmental variables measured in the study area reflects gradients in physical conditions and refers to a transition from a “high energy” and a “stressed” sandy shallow environment to a deeper and more stable muddy one, attributed mainly to natural forcing like storms and winds, strength of currents and turbidity. This is further supported by the low sediment POC concentrations measured in the study area, which are well below the lower critical point (<10 mg/g) corresponding to coastal areas without organic loading and associated stressors (Hyland et al. 2005). This environmental setup corresponds to the erosion, high combustion and deposition sedimentary coastal areas exposed to the wave action described by Graf (1992).

The relatively low concentrations of heavy metals, even in front of the industrialised part of the coastal area of Heraklion Bay, reveal the significant role of coastal marine processes (e.g. wave activity, shelf water circulation) that serve the ‘self cleaning’ of the local coastal system. Trace metals are transferred along with the fine-grained sediment seawards to the wave breaking zone, where they become

Table 6 Values of Spearman's correlation coefficient between the scores of PCA analysis of the three sub-areas (I, II and III) and the normalized environmental variables (significant level $\rho < 0.001$)

Variables	PC axis (sub-area I)	ρ	PC axis (sub-area II)	ρ	PC axis (sub-area III)	ρ
Depth (m)	1	-0.8	1	+0.9	1	-0.8
Eh (mV)	1	+0.8	-	-	1	+0.8
Mud (%)	1	-0.9	1	+0.9	1	-0.9
CPE ($\mu\text{g/g}$)	-	-	-	-	-	-
POC (mg/g)	1	-0.8	-	-	1	-0.8
Cu (ppm)	3	+0.7	-	-	2	-0.6
Hg (ppm)	-	-	1	-0.8	-	-
Pb (ppm)	2	-0.8	2	-0.9	-	-
Cd (ppm)	2	-0.9	-	-	-	-
Zn (ppm)	-	-	2	-0.8	2	-0.6

Eh redox potential; mud = silt + clay; *CPE* chloroplastic pigment equivalent, *POC* particulate organic carbon

subject to the eastward prevailing (offshore) shelf-water circulation pattern; the latter seems to act both as a natural barrier to their further seaward dispersion and as a 'conduit' that transports eastwards the suspensates, including trace metals. The anti-cyclone gyre in Malia Bay by entrapping the advected material presents locally low trace metal concentrations.

The heavy metal concentrations in the surficial shelf sediment along the northern coast of Heraklion Prefecture are not considered to be dangerous for human health, as

they are lower than the standard WHO values, with only a few localised exceptions.

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Appendix I

Table 7

Table 7 The water depth (m), redox potential (Eh in mV), percentage of mud (silt + clay), chloroplastic pigment equivalent (CPE in $\mu\text{g/g}$), particulate organic carbon (POC in mg/g) and trace metal concentrations (ppm) of each surficial sediment sample

Stations	Depth (m)	Eh (mV)	Mud (%)	CPE ($\mu\text{g/g}$)	POC (mg/g)	Cu (ppm)	Hg (ppm)	Pb (ppm)	Cd (ppm)	Zn (ppm)
1	10	326	3.91	1.1668	0.236	25.35	0.035	10.4	0.035	33.04
2	20	369	1.31	0.6864	0.323	28.83	0.04	12.17	0.077	38.53
3	30	161	13.91	6.8542	2.043	117.86	0.066	30.91	0.112	88.26
4	40	98	82.5	8.6745	7.042	49.12	0.052	29.81	0.183	59.04
5	10	241	11.1	1.4906	5.17	56.76	0.053	21.03	0.039	52.66
6	20	39	18.36	2.6793	7.002	126.1	0.043	20.19	0.063	77.48
7	30	54	48.22	6.0337	6.61	61.96	0.053	25.12	0.102	81.35
8	40	160	73.06	13.0231	7.002	53.71	0.067	27.41	0.278	80.09
9	20	49	44.58	34.931	10.126	89.91	0.044	42.25	0.095	74.82
10	40	48	68.16	13.9625	8.788	39.03	0.066	30.67	0.327	68.33
11	10	389	5.22	1.6582	3.098	16.68	0.055	14.77	0.061	31.07
12	20	134	8.25	3.0608	3.163	21.23	0.045	17.94	0.059	35.6
13	30	126	54.64	4.7789	3.39	99.18	0.067	26.13	0.085	102.81
14	40	64	86.18	2.1341	6.244	74.51	0.043	27.3	0.081	114.94
15	70	86	90.11	5.8318	4.101	28.37	0.033	28.05	0.047	89.52
16	10	353	2.25	1.0536	0.021	23.14	0.033	10.42	0.083	41.27
17	20	332	4.35	1.2202	0.797	31.05	0.131	12.16	0.097	48.54
18	30	70	87.21	3.3171	5.527	84.6	0.089	39.99	0.092	99.35
19	40	87	73.79	4.9051	6.365	59.41	0.056	40.73	0.079	101.52
20	70	111	96.78	9.7602	6.169	27.82	0.043	36.22	0.053	74.99
21	10	354	2.7	1.3468	0.052	13.65	0.046	29.33	0.286	29.63

Table 7 continued

Stations	Depth (m)	Eh (mV)	Mud (%)	CPE ($\mu\text{g/g}$)	POC (mg/g)	Cu (ppm)	Hg (ppm)	Pb (ppm)	Cd (ppm)	Zn (ppm)
22	20	265	9.13	2.4103	0.709	21.66	0.077	37.25	0.377	35.9
23	30	78	38	15.0321	2.286	70.31	0.066	47.17	0.607	77.02
24	40	84	72.77	2.1341	4.538	51.19	0.074	45.62	0.579	79.64
25	10	91	7.07	7.5845	1.794	52.52	0.208	48.23	1.860	99.23
26	20	66	7.84	3.3368	0.992	35.88	0.08	45.56	0.790	82.07
27	30	93	6.76	4.3545	0.716	37.12	0.069	63	1.470	125.32
28	40	184	9.54	8.3516	2.546	29.72	0.046	61.09	1.390	110.76
29	70	74	97.42	2.8765	5.969	23.43	0.034	57.92	0.831	61.7
30	30	337	3.11	4.5609	0.751	33.72	0.074	58.06	1.219	52.17
31	40	184	10.66	5.3295	1.606	25.1	0.084	51.7	0.793	81.33
32	20	114	17.55	4.0491	0.259	29.72	0.057	47.69	0.608	66.24
33	30	104	11.69	6.6967	3.079	31.25	0.058	50.65	0.831	83.52
34	40	119	17.71	7.1199	3.329	27.91	0.045	42.71	0.948	92.4
35	70	174	83.21	12.254	3.998	19.12	0.064	35.04	0.741	58.34
36	10	601	2.56	1.151	1.762	23.47	2.037	27.51	0.967	47.53
37	10	399	2.55	1.6462	0.463	19.81	1.922	30.17	0.872	42.58
38	20	274	4.42	1.8245	2.331	20.53	1.416	29.44	0.601	39.84
39	30	174	29.63	6.4773	3.609	35.74	1.009	33.26	0.679	50.01
40	40	154	74.33	7.1661	5.319	28.07	0.107	30.87	0.500	45.77
41	70	206	90.09	5.5101	3.463	17.06	0.036	26.62	0.106	33.08
42	10	406	4.29	3.9667	0.031	14.11	0.09	14.46	0.074	22.54
43	20	584	25.81	4.0146	7.54	19.23	0.113	19.49	0.043	25.39
44	30	354	28.72	4.6057	3.919	37.3	0.113	22.6	0.076	41.5
45	40	120	58.87	5.5463	3.295	24.19	0.06	20.31	0.058	46.51
46	70	94	58.67	2.4538	2.309	16.52	0.06	18.55	0.036	39.08
47	10	419	2.29	1.4584	0.075	17.13	1.161	11.89	0.093	25.71
48	20	408	4.18	5.7024	3.417	21.61	0.622	15.37	0.068	34.1
49	30	151	50.07	5.4029	5.408	28.12	0.432	24.18	0.084	39.49
50	40	174	47.22	4.3984	3.838	23.09	0.451	22.61	0.063	45.14
51	70	434	57.00	5.0797	2.401	18.76	0.1	20.88	0.041	35.18
52	10	382	1.88	1.7856	0.519	34.08	0.135	13.85	0.104	20.31
53	20	284	2.79	1.7254	1.134	29.13	0.181	12.75	0.115	18.5
54	30	164	32.47	4.7224	2.799	30.21	0.699	19.3	0.427	25.06
55	40	177	45.64	5.614	3.322	28.14	0.33	17.42	0.389	17.11
56	30	254	31.47	5.1285	3.697	5.21	0.251	10.31	0.101	10.09
57	40	109	34.08	3.9468	3.729	4.03	0.043	11.08	0.1	15.62
58	70	109	74.96	7.3332	2.812	3.01	0.035	17.22	0.075	7.99
59	20	375	1.96	1.0159	0.945	–	–	–	–	–
60	30	397	1.56	0.793	3.392	3.11	0.194	9.87	0.097	8.29
61	40	351	23.01	3.7788	2.422	3.05	0.077	10.45	0.083	11.94
62	70	249	59.27	1.9778	4.281	2.07	0.034	14.51	0.044	6.89
63	10	373	3.1	1.8238	0.901	5.46	0.133	17.48	0.058	15.98
64	20	178	6.48	1.7218	0.135	6.46	0.172	17.98	0.071	22.31
65	30	155	28.51	2.1075	4.478	29.5	0.209	25.79	0.086	28.03
66	40	79	53.32	6.2862	5.421	17.02	0.076	23.16	0.075	32.4
67	70	214	45.86	9.8025	3.883	11.14	0.085	19.73	0.037	14.17
68	20	203	8.71	2.6766	2.245	5.8	0.246	15.48	0.065	12.22
69	30	136	35.06	–	3.927	16.91	0.162	18.06	0.077	20.01

Table 7 continued

Stations	Depth (m)	Eh (mV)	Mud (%)	CPE ($\mu\text{g/g}$)	POC (mg/g)	Cu (ppm)	Hg (ppm)	Pb (ppm)	Cd (ppm)	Zn (ppm)
70	40	148	63.02	8.5538	5.343	13.42	0.117	16.98	0.061	19.44
71	10	331	3.26	1.5554	1.941	7.89	0.162	11.51	0.061	12.86
72	20	169	4.35	1.9445	2.513	10.34	0.131	9.86	0.058	10.15
73	30	204	25.84	4.4621	3.058	19.32	0.079	12.73	0.05	25.32
74	40	154	74.6	3.3305	2.065	16.09	0.066	13.08	0.064	27.12
75	70	72	85.08	5.3721	4.214	9.17	0.035	10.71	–	12.68
76	10	381	2.6	1.9887	1.607	4.26	0.103	9.58	0.055	8.79
77	20	312	3.61	3.1175	1.296	3.38	0.087	10.07	0.079	13.06
78	30	232	6.6	8.1271	1.661	5.12	0.105	16.92	0.083	19.17
79	40	266	42.01	6.8139	5.323	4.23	0.117	15.49	0.079	15.86
80	20	154	1.75	2.5836	0.116	4.01	0.098	11.53	0.049	12.54
81	30	380	14.18	3.1467	2.629	6.27	0.076	16.54	0.071	15.06
82	40	170	92.86	2.3258	3.332	3.92	0.046	14.92	0.08	17.23
83	70	173	96.3	8.1208	3.429	3.15	0.055	10.39	–	13.86
84	10	279	6.5	0.9433	0.103	4.16	0.159	8.33	0.049	9.24
85	20	488	1.54	–	1.062	4.3	0.207	8.26	0.068	11.78
86	30	340	9.46	19.74	1.897	11.18	0.115	17.42	0.08	17.79
87	40	114	20.15	12.8698	0.999	9.42	0.034	15.79	0.091	15.44
88	70	53	91.81	3.4517	3.969	3.04	0.046	12.99	0.049	12.38

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