Sediment Pollution by Heavy Metals in the Strymonikos and Ierissos Gulfs, North Aegean Sea, Greece

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SEDIMENT POLLUTION BY HEAVY METALS IN THE STRYMONIKOS AND IERISSOS GULFS, NORTH AEGEAN SEA, GREECE

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Abstract. Surface sediment samples from Strymonikos and Ierissos Gulfs were analyzed for Cu, Pb, Zn, Cr and Ni. The results showed that the sediment of Ierissos Gulf is more polluted with Cu, Pb, and Zn as compared to that of Strymonikos Gulf. The benthal area located off the load-out facility of the mining operations in the town of Stratoni, in Ierissos Gulf is established as the most polluted region. The distribution of Cr and Ni in both gulfs indicates the natural origin of these metals with the weathering of Strymon River and of other smaller rivers rocks being responsible for their enrichment.

Keywords: Greece, heavy metals, sediment pollution, Strymonikos and Ierissos Gulfs

1. Introduction

Marine sediments are a major repository of heavy metals in coastal areas (Solomons and Förstner, 1984). Therefore, sediment analyses play an important role in the quality assessment of the marine environment in so far as metal pollution is concerned.

The coastal marine ecosystems of Strymonikos and Ierissos Gulfs (Figure 1) are commercial fishing grounds (trawling is not allowed in the Ierissos Gulf) and are adjacent to areas of important industrial and agricultural activities.

In the north part of the Strymonikos Gulf, the Strymon River discharges its waters and it is one very important source of pollution for the gulf, because it carries agricultural wastes originating from activities in the plain of Serres valley, domestic sewage as well as effluents from the industrial estates that are located along its banks, in Greece as well as in Bulgaria. The catchment area of the Strymon River is 17 130 km^2, approximately 63% of which belongs to Bulgaria. Because its drainage basin contains maphic and ultramaphic rocks (ophiolites) rich in Cr and Ni, the weathering products contain elevated concentrations of such metals (Varnavas, 1986, 1989). The effect of the river drainage basin mineralogy on the geochemistry of the sediments of Strymonikos Gulf has been suggested in previous investigations in the area (Konispoliatis, 1984; Sakellariadou, 1993). Furthermore,

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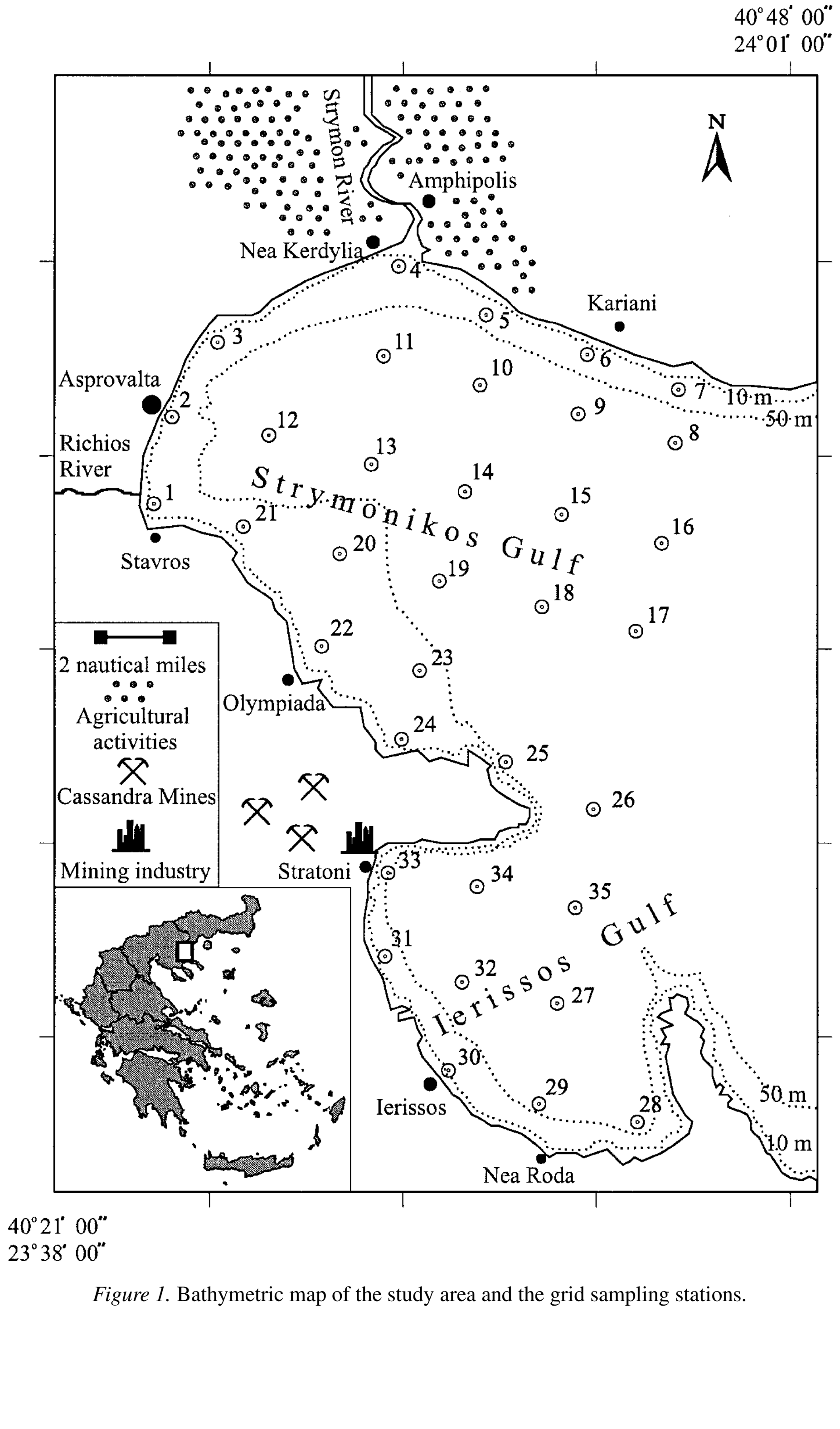

Figure 1. Bathymetric map of the study area and the grid sampling stations.

in the northwest part of the Strymonikos Gulf, Richios River flows into the gulf and it carries agricultural wastes from the Laggadas valley.

However, the most important source of environmental problems concern in the study area due to the Cassandra Mines, located in the northeast part of Chalkidiki Prefecture of north Greece, which exploit the mixed sulfide ores of north Greece. The mixed sulfide ores contain mainly galenite (blue lead) for Pb and Ag exploitation and sphalerite (zinc blende) for Zn exploitation. These mines have been exploited for more than 2500 yr, since the era of Alexander the Great. In the recent past, the mining operations included dispersal of mine tailings into the nearshore marine environment in the vicinity of Stratoni, in the Ierissos Gulf. In 1985 the Cassandra Mines were closed, but were opened again in 1995. During the 1980s, preliminary surveys of heavy metal pollution were carried out in the north Aegean Sea including Strymonikos and Ierissos Gulfs (Sakellariadou, 1987, 1991; Konispoliatis and Perissoratis, 1987). A significant enrichment of Zn and Pb was identified in surface sediments of the northwest Ierissos Gulf. Surface marine sediment values from adjacent areas in north Aegean denote this enrichment. However, heavy metal pollution in the sediments of both gulfs has not been studied in recent years and particularly not since the reopening of the Cassandra Mines. Latest investigations on the abundance and the taxonomic diversities of some marine species as well as on the nutrient concentrations in both gulfs still shows the presence of pollution in Ierissos Gulf. The qualitative study of the marine benthic macroflora shows three phanerogames (*Cymodocea nodosa, Posidonia oceanica, Zostera noltii*) in Strymonikos Gulf but only one (*Posidonia oceanica*) in Ierissos Gulf (Lazaridou *et al*., 1999). The qualitative and quantitative study of the marine benthic macrofauna shows higher values in Strymonikos Gulf for the number and the quantity of species as compared to those measured in Ierissos Gulf (Vafidis and Dailianis, 1999). Seawater samples from inshore stations of the Strymonikos Gulf showed higher concentrations of nutrients and a greater abundance of chlorophyll, than those from the heavy metal polluted region of Ierissos Gulf (Stamatis *et al*., 2001; Stamatis and Ioannidou, 1999).

The aim of this study was (1) to determine the present heavy metal pollution state in the two gulfs, (2) to examine whether significant changes have occurred over the last few years and (3) to compare the results from this area with those available from other Greek and Mediterranean areas. To that end, metal concentrations in surface sediments were collected in a dense network of 35 sampling stations.

2. Materials and Methods

2.1. Samples

Samples of marine surface sediments were collected from the Strymonikos and Ierissos Gulfs at 35 grid stations (Figure 1). This grid arrangement was designed so as to enable the determination of the spatial distribution of heavy metals within the whole geographic area studied. Tidal ranges in the study area vary between 18–20 cm during spring tides and between 9–10 cm during neap tides. The depth of the sampling stations shows Table I.

Sediment samples were collected in the time interval from 19 to 22 February 1998 using a Van Veen stainless steel grab (20 × 20 cm). Samples were taken with a plastic spoon (5 mL) from the top and middle of the grab to detect the recent heavy metal pollution. The sediment samples were immediately placed at 0 °C, on board. Subsequently, the samples were stored at –28 °C in plastic vessels, cleansed with nitric acid (1 N) for 1 hr and rinsed thoroughly with bi-distilled water.

2.2. Digestion

The sediment samples were defrosted at room temperature, dried at 50 °C up to a constant weight, and, after cooling to room temperature, ground in a mortar to a fine powder. Then, three subsamples each of 1 g of the gut homogenized sediment sample were digested with 10 mL nitric acid (2 N) in an autoclave-3 (BOO7-8486, Perkin Elmer) at 140 °C, for 1 hr. It is known that such digestion does not give the total metal concentration but only up to 80% of the total element content for metals like Ni and Cr.

2.3. Analysis

Concentrations of Cu, Pb, Zn, Cr and Ni were determined in the filtered and diluted solutions, in triplicate, by atomic absorption spectroscopy (AAS) using a PC controlled Perkin-Elmer 5100ZL spectrometer, equipped with a THGA graphite furnace, ZEEMAN background correction and AS-70 auto sampler. The quantity assurance of the analytical results was controlled with the use of ‘BCSS-1 marine sediment’ as Reference Material.

2.4. Statistics

For the interpretation of the data of the chemical analysis the following methodology was used:

(1) independent *t*-test on the metal concentrations from the two gulfs separately was performed;

TABLE I

Depth of the sampling stations, metal mean concentrations and *t*-test results in the study area

Station	Depth (m)	Concentration (ppm)				
		Cu	Pb	Zn	Cr	Ni
1	15.0	16.2	39.2	90.0	136.1	51.5
2	15.0	10.4	23.4	53.2	103.0	35.2
3	25.0	29.3	44.7	94.9	178.7	64.0
4	14.5	42.0	74.3	135.6	187.7	71.9
5	31.5	34.7	78.9	118.7	150.1	54.2
6	15.0	35.9	92.5	127.6	183.6	69.3
7	29.0	7.0	29.7	43.2	53.5	20.5
8	67.7	28.3	82.3	107.6	148.0	57.4
9	68.5	35.2	91.7	126.4	178.2	69.2
10	65.5	36.4	106.7	146.0	203.3	72.1
11	53.0	36.5	108.1	144.8	197.0	68.0
12	54.0	36.6	123.2	147.5	201.9	71.3
13	55.0	36.7	124.2	153.5	213.0	74.5
14	54.0	7.7	79.3	55.3	74.3	26.5
15	69.0	31.0	116.4	135.2	192.2	67.8
16	77.0	30.1	122.6	136.9	188.1	66.6
17	78.0	51.2	130.5	159.0	185.0	70.1
18	69.0	30.3	124.8	134.2	187.3	66.3
19	53.0	29.4	124.3	90.7	116.8	47.2
20	40.0	5.3	57.2	50.2	62.3	18.9
21	42.0	34.5	115.9	141.3	196.8	70.5
22	20.0	10.7	115.8	94.5	75.6	30.0
23	42.0	26.8	111.6	127.8	170.3	59.7
24	16.0	40.8	127.8	128.0	105.7	35.6
25	32.0	8.1	32.4	24.3	29.2	9.1
26	68.0	24.4	116.7	124.6	150.6	53.2
27	73.0	50.5	440.2	387.5	242.3	95.0
28	59.0	56.6	511.7	486.5	264.1	100.4
29	60.0	48.7	226.5	260.8	134.9	49.6
30	12.5	0.0	60.4	39.6	17.1	2.1
31	25.0	179.1	1558.1	889.4	364.2	144.3
32	66.0	70.0	683.5	595.0	269.1	112.7
33	14.0	205.9	2233.1	926.8	173.5	32.0
34	76.0	58.4	493.5	433.4	237.5	85.0
35	33.5	8.3	52.9	57.1	53.6	14.4
t-test – Strymonikos Gulf (Stations 1–26)						
Mean		27.1	92.1	111.2	148.8	53.9
t-test – Ierissos Gulf (Stations 27–35)						
Mean		75.4	695.5	452.9	195.1	70.6

(2) cluster analysis among sampling stations for the individual metals was applied; dendrograms were created for each metal on logarithmic transformed data (Davis, 1986), with the Average Linkage method and the similarity measure was the Euclidean distance (Andreev and Simeonov, 1986);
(3) pollution ratio for each metal and for its subarea was calculated; Pollution ratio is the average metal concentration of a region divided by the metal concentration of the subarea I, which is the area with the lowest heavy metal content;
(4) maps of the areal distribution for each metal were drawn;
(5) interelement correlation coefficients (r), by $p < 0.05$, were calculated.

Statistics were performed using commercial available PC-software.

3. Results and Discussion

The results of the chemical analyses given by the metal mean concentrations are shown in Table I.

Independent *t*-test applied to the Cu, Pb and Zn concentrations from the two gulfs separately, shows significant differences for their mean values ($p < 0.05$). In contrast the mean values of Cr and Ni are not significantly different ($p < 0.05$). These *t*-test results presented as the mean values for the two gulfs separately are also shown in Table I. Cluster analysis for the individual metals identified four principal groups, corresponding to the subareas I, II, III and IV. The sampling stations included in the various subareas of the study region are given in Table II.

Mean metal subareal content can be ranked by abundance in the marine sediments of the study area as follows: sub I < sub II < sub III < sub IV. Subarea I is valid as reference.

3.1. Spatial distribution of the metals – comparison of the concentrations

The spatial distribution of the metals is shown for each metal separately, in the Figures 2a–e. Table III shows the metal levels (mean, range) and the pollution ratio in the various subareas I, II, III and IV. Table IV shows the highest level of the metal concentrations in surface sediments in various east Mediterranean coastal ecosystems. Despite the difficulties involved in a comparison of the metal concentrations in the sediments of Mediterranean gulfs, lagoons or coasts, due to the wide assortment of the analytical procedures used, a comparison of their highest levels is only feasible and indicative when the relative degree of metal extraction of each digestion method is taken into account (Table IV).

TABLE II
Sampling stations included in various subareas of the study region

Area	Cu	Pb	Zn	Cr	Ni
I	1, 2, 7, 14, 20, 22, 25, 30, 35	1–26, 30, 35	1–26, 30, 35	1, 2, 5, 7, 8, 14, 19, 20, 22–26, 29, 30, 35	25, 30, 35
II	3–6, 8–13, 15, 16, 18, 19, 21, 23, 26	29	29	3, 4, 6, 9, 10–13, 15–18, 21, 33	2, 7, 14, 19, 20 22, 24, 29, 33
III	17, 24, 7–29, 32, 34	27, 28, 32, 34	27, 28, 32, 34	27, 28, 32, 34	1, 3–6, 8–13, 15–18, 21, 23, 26–28, 34
IV	31, 33	31, 33	31, 33	31	31, 32

TABLE III
Levels of heavy metals (ppm) in various subareas of Strymonikos and Ierissos Gulfs

Area		Cu	Pb	Zn	Cr	Ni
I	Mean	8.2	61.9	99.7	105.7	8.5
	Range	0.0–16.2	23.4–92.5	43.2–135.6	53.5–150.6	2.1–14.4
II	Mean	34.9	226.5	260.8	185.5	32.8
	Range	28.3–42.0	–	–	170.3–196.8	18.9–49.6
	Pollution ratio	4.6	3.7	2.6	1.8	3.9
III	Mean	53.7	532.2	475.6	256.9	69.4
	Range	40.8–70.0	440.2–683.4	387.5–595.0	237.5–269.1	51.5–100.4
	Pollution ratio	7.0	8.6	4.8	2.4	8.2
IV	Mean	192.5	1895.6	908.1	364.2	128.5
	Range	179.1–205.9	1558.1–2233.1	889.4–926.8	–	112.7–144.3
	Pollution ratio	23.5	30.6	9.1	3.5	15.1

TABLE IV

Highest level of the metal concentrations (ppm) in surface sediments in various east Mediterranean coastal ecosystems

Area	Cu	Pb	Zn	Cr	Ni	Digestion Method	Reference
Strymonikos Gulf (Greece)	51.2	130.5	159.0	213.0	74.5	2 N HNO_3, autoclave	Present study
Strymonikos Gulf (Greece)	128	313	236	–	110	Total	(a)
Ierissos Gulf (Greece)	205.9	2233.1	926.8	364.2	144.3	2 N HNO_3, autoclave	Present study
Ierissos Gulf (Greece)	75	586	1454	–	–	Total	(b)
Kavala Gulf (Greece)	1.3	12.6	15.9	–	–	2 N HNO_3	(c)
Thermaikos Gulf (Greece)	69	268	560	299	–	HNO_3 conc.	(d)
Saronikos Gulf (Greece)	1000	–	720	–	–	2 N HCl	(e)
Pagassitikos Gulf (Greece)	32	34	74	395	–	2 N HCl	(f)
Kerkyra Strait (Greece)	30	24	94	257	192	2 N HCl	(g)
Amvrakikos Gulf (Greece)	31	21	80	177	188	2 N HCl	(g)
Navarino Gulf (Greece)	32	28	81	251	123	2 N HCl	(g)
Messolonghi Lagoon (Greece)	30	23	109	246	112	2 N HCl	(g)
Venice Gulf (Italy)	44.2	49.9	446.0	–	–	Total	(h)
Venice Lagoon (Italy)	463	278	5930	–	–	Total	(i)
Naples coast (Italy)	95	775	1600	–	–	Total	(j)
Croatia coast (Croatia)	550	1275	1300	–	–	Total	(k)
Average deep-sea clays	250	80	–	90	–	Total	(l)

(a) Konispoliatis, 1984; (b) Sakellariadou, 1987; (c) Fytianos and Vasilikiotis, 1982; (d) Voutsinou-Taliadouri and Satsmadjis, 1983; (e) Voutsinou-Taliadouri, 1981; (f) Voutsinou-Taliadouri, 1984; (g) Voutsinou-Taliadouri, 1998; (h) Angela *et al.*, 1980; (i) Pavoni *et al.*, 1987; (j) Sharp and Nardi, 1987; (k) Stegnar *et al.*; 1980; (l) Turekian and Wedepohl, 1961.

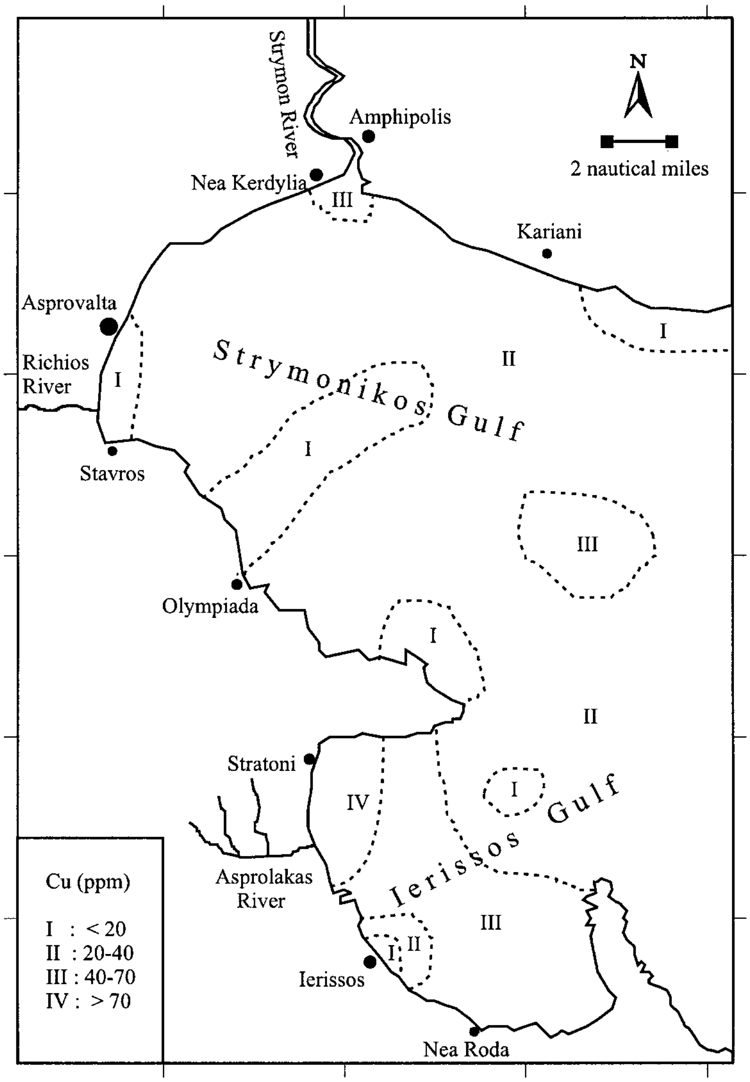

Figure 2a. Areal distribution of Cu.

3.1.1. *Copper*

The spatial distribution of Cu is shown in Figure 2a. The highest Cu mean subareal content (192.5 ppm, Table III) was found at the load out pier of the mining operations in the Ierissos Gulf (area IV). The pollution ratio of this area has its highest value as well (23.5, Table III). That shows that Cu main source is effluent from the Cassandra Mines. The highest score value for Cu was found at station 33 (205.9 ppm) and it is higher compared to the highest level for Cu from a previous study (75 ppm, Table IV) (Sakellariadou, 1987). It is also higher compared with Cu levels measured in other coastal ecosystems like Strymonikos, Kavala,

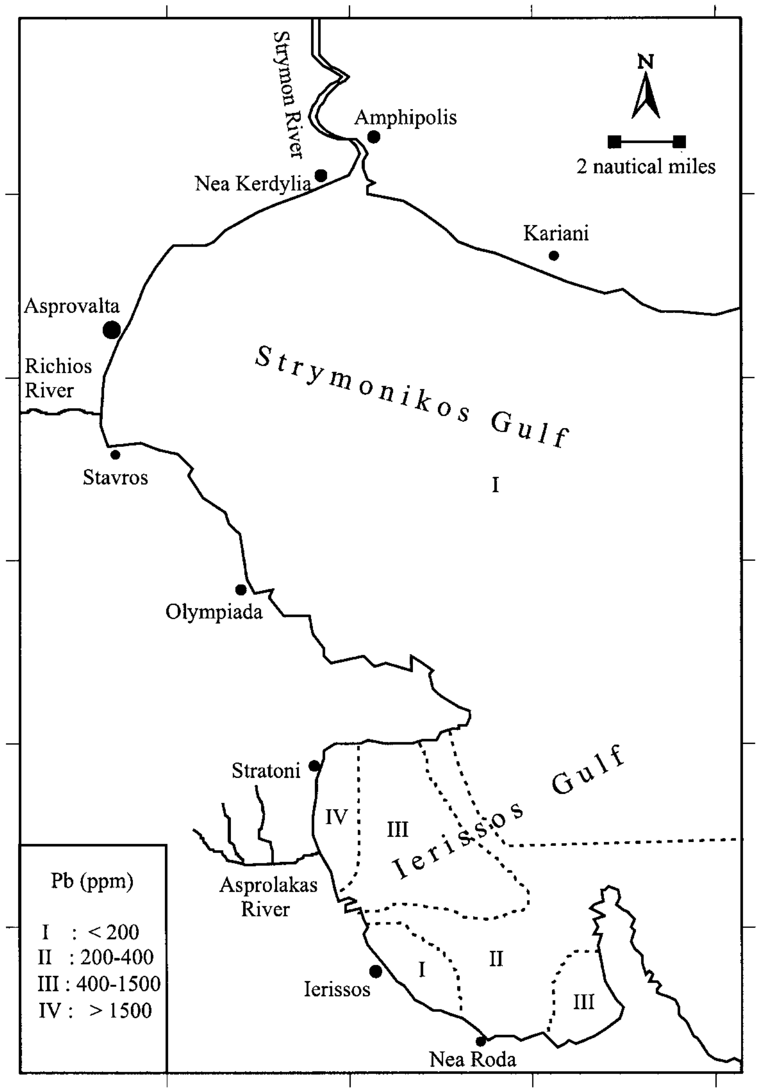

Figure 2b. Areal distribution of Pb.

Thermaikos, Pagassitikos, Amvrakikos, Navarino and Venice Gulf, Kerkyra Strait, Messolonghi Lagoon and Naples coast, (Table IV). Cu highest score from this study is lower than that found in Saronikos Gulf, Venice Lagoon and Croatia coast (Table IV). The higher metal content in Saronikos Gulf compared to that measured in our study area can be coherent with the higher quantities of organic mutter in the gulf, caused mainly from untreated municipal and industrial effluents of Athens and the surrounding areas. The influence of organic mutter for the sediment enrichment with metals is well known. The remaining area of the Ierissos Gulf (excepting an

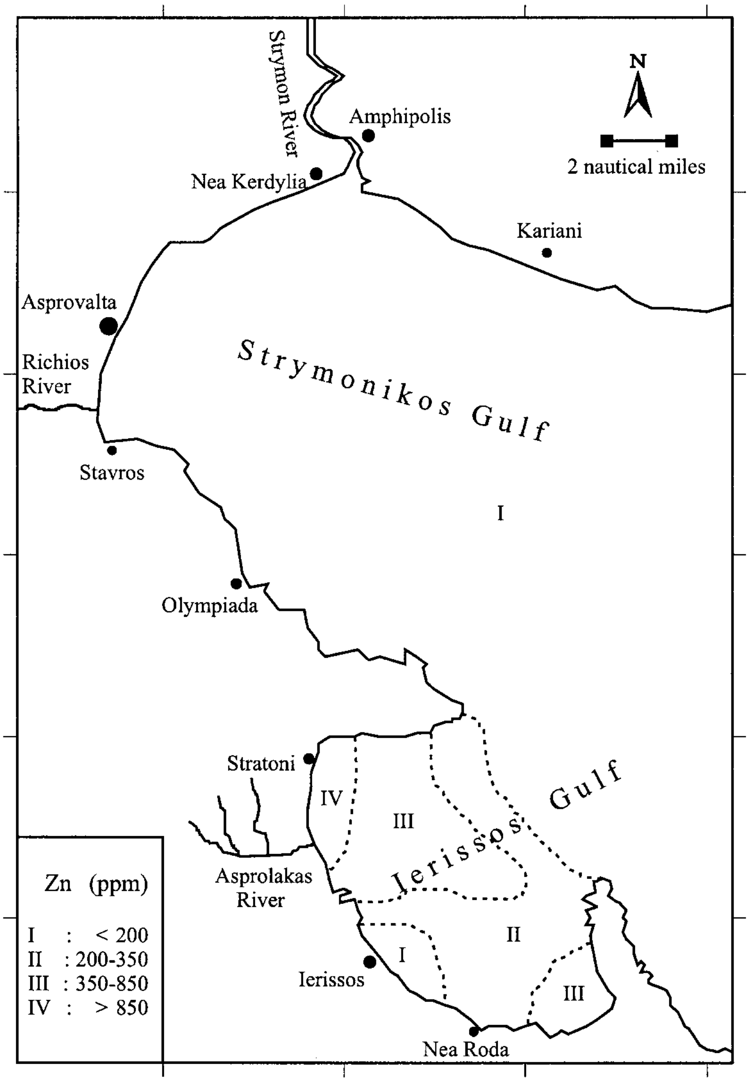

Figure 2c. Areal distribution of Zn.

small benthal region in the town of Ierissos), the area in the mouth of Strymon River, as well as a small region at offshore Strymonikos Gulf (station 17), forms the area III (Figure 2a) with metal levels varying from 40.8 to 70.0 ppm and pollution ratio 7.0 (Table III). Except area I, which is valid for Cu as reference area (<16.2 ppm) the rest of the studied region is area II (Figure 2a), which shows moderate Cu concentrations (28.3–42.0 ppm) and pollution ratio 4.6.

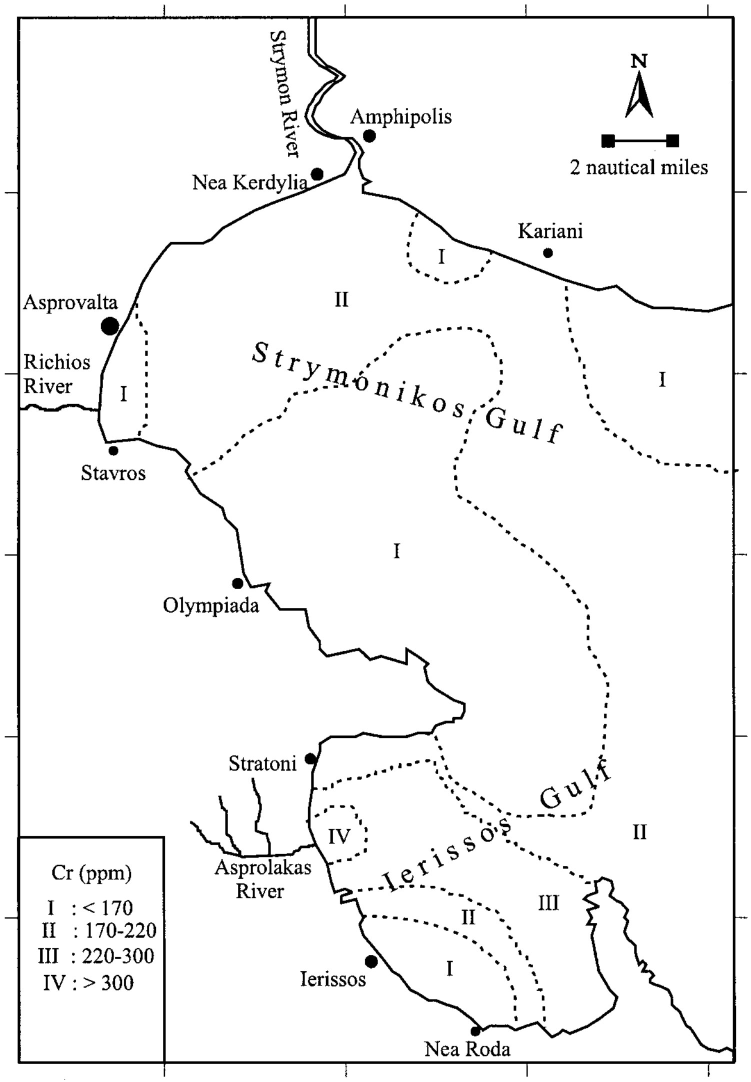

Figure 2d. Areal distribution of Cr.

3.1.2. *Lead and zinc*

Lead and Zn have very similar spatial distribution (Figures 2b and c): their highest subareal mean values (1895.6 and 908.1 ppm, Table III) were found at the load out pier of the mining operations (area IV), then at the neighbouring areas III (532.2 and 475.6 ppm) and II (226.5 and 260.8 ppm). The stations 30 and 35 of Ierissos Gulf as well as the stations of Strymonikos Gulf form area I with the lowest content of Pb and Zn (61.9 and 99.7 ppm, respectively). The spatial distribution of Pb and Zn indicates that their main source is effluent from the mines, where the pollution

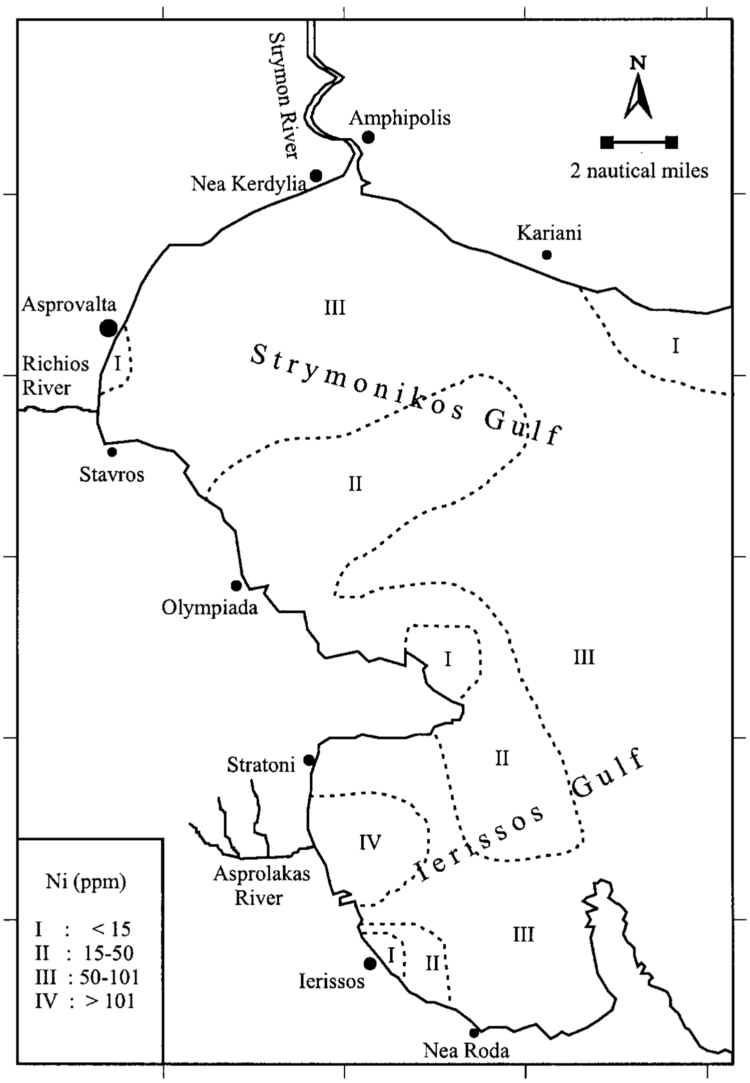

Figure 2e. Areal distribution of Ni.

ratios for these metals have their highest values, namely 30.6 and 9.1, respectively (Table III).

The highest score value for Pb was found at station 33 (2233.1 ppm) in the Ierissos Gulf. This score is higher compared to that detected for Pb (586 ppm, Table IV) in a previous study in the area (Sakellariadou, 1987). Lead highest score from this study is also higher compared with those found for Pb in other Greek and Mediterranean coastal regions (Table IV).

Like Pb, the highest score for Zn was found at station 33 (926.8 ppm) in the Ierissos Gulf. This score is lower compared to that measured for Zn (1454 ppm,

Table IV) in a previous study in the area (Sakelariadou, 1987). It is also lower compared with the highest scores for Zn, found in Venice Lagoon (5930 ppm) Naples coast (1600 ppm) and Croatia coast (1300 ppm). Zinc highest score from this study is higher compared with those found in other Greek and Mediterranean coastal ecosystems, like Strymonikos, Kavala, Thermaikos, Saronikos, Pagassitikos, Amvrakikos, Navarino and Venice Gulf, Kerkyra Strait and Messolonghi Lagoon (Table IV).

3.1.3. *Chromium and nickel*

Chromium and Ni have similar spatial distribution (Figures 2d and e) which is completely different from that of Cu, Pb and Zn. The highest mean subareal content of Cr (364.2 ppm) and Ni (128.5 ppm) was detected at area IV in the Ierissos Gulf. Area IV seems to be affected slightly effluent from the small Asprolakas River with drainage basin rich in Cr and Ni (Ioannidou *et al.*, 1999). The pollution ratios in area IV were 3.5 and 15.1 for Cr and Ni, respectively (Table III).

The highest score of Cr was found at station 31 (364.2 ppm) in the Ierissos Gulf and it is higher compared to the highest values for Cr found in other coastal ecosystems in Greece, like Strymonikos (present study), Thermaikos, Amvrakikos, Navarino Gulfs, Kerkyra Strait and Messolonghi Lagoon and lower to the highest Cr content from Pagassitikos Gulf. High Cr concentrations were detected as well at the stations 27 (242.3 ppm), 28 (264.1 ppm), 32 (269.1 ppm) and 34 (237.5 ppm). The above stations forms area III (Figure 2d) with pollution ratio 2.4 (Table III). The remaining of the study region is divided by ca. 50 % to area II with moderate Cr content (170–220 ppm, pollution ratio = 1.8) and by ca. 50% to area I with the lowest Cr content (<170 ppm). Like Cr, the highest score of Ni was measured at station 31 (144.3 ppm). This score is higher compared to the highest values for Ni found in Navarino Gulf and Messolonghi Lagoon and lower to the highest Ni content from Kerkyra Strait and Amvrakikos Gulf (Table IV). High Ni concentrations were measured as well at station 32 (112.7 ppm) (area IV) and at the stations of the area III (51.5–100.4 ppm). Areas II and I cover the rest of the study area (Figure 2e) and shows moderate (18.9–49.6 ppm) and low (<15 ppm) Ni concentrations, respectively.

Area III for Ni and area II for Cr seem result mainly from the effluent of Strymon River. It is highly probable that Cr and Ni at both gulfs are derived mostly from the weathering of maphic and ultramaphic rocks being traversed by Strymon River and other smaller rivers (e.g. Asprolakas River). Chromium and Ni concentrations appear to increase at offshore and to decrease at central Strymonikos Gulf, forming sediment zones of lower concentrations. It has been suggested that the tidal water circulation forms an anti-clockwise eddy close to the mouth of Strymon River (Sylaios, 1999). This may explain why higher metal concentrations were observed at offshore areas with lower metal concentrations measured at central areas of the Strymonikos Gulf.

TABLE V

Interelement correlations for surface sediments from the whole study area

	Cu	Cr	Pb	Zn	Ni
Cu	1				
Cr	0.546	1			
Pb	0.954*	0.468	1		
Zn	0.920*	0.667	0.946*	1	
Ni	0.425	0.976*	0.341	0.577	1

* High significance by $p < 0.05$.

3.2. Interelement correlation coefficients (r)

Zinc concentration levels showed highly significant correlation with those of Pb concentration levels (r = 0.946, $p < 0.05$), (Table V). Copper values are highly significant correlated with Zn (r = 0.920, $p < 0.05$) and Pb (r = 0.954, $p < 0.05$) values as well. These three metals come mostly from the effluent due to mill operations in Stratoni (station 33) affecting mainly the sediments of Ierissos rather than those of the Strymonikos Gulf. Ni concentration levels correlate highly with Cr concentration levels (r = 0.976, $p < 0.05$). Ni and Cr are transported into the gulfs more as weathering product from river runoff.

4. Conclusions

The most polluted area for Pb, Zn and Cu in both gulfs is the benthal area located near the load-out facility of the mining operations ('flotation') in Stratoni Bay. In particular, the inshore northwest region of Ierissos Gulf is one of the most polluted coastal ecosystems of the east Mediterranean by Pb and Zn. Chromium and Ni in both gulfs are probably a result of the weathering action of Strymon River, as well as of other smaller rivers, that flow into these gulfs. The result of the present work demonstrate in Ierissos Gulf higher Cu and Pb content as compared to previous study. In contrast, the Zn content measured here is lower than that of previous study. In Strymonikos Gulf the present study demonstrates decrease in the content of Cu, Pb, Zn and Ni than that measured in previous study. The grid sampling showed an expected decreasing abundance of the metals in the sediment offshore of the Ierissos Gulf. On the contrary, in the Strymonikos Gulf the metal content appears to decrease in central areas and it seems to increase offshore, probably as a result of the tidal water circulation.

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