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Study on the behavior of the heavy metals Cu, Cr, Ni, Zn, Fe, Mn and ^{137}Cs in an estuarine ecosystem using *Mytilus galloprovincialis* as a bioindicator species: the case of Thermaikos gulf, Greece

Vassiliki-Angelique Catsiki [a,*], H. Florou [b]

[a] *Hellenic Centre for Marine Research, Mavro Lithari, 46.7 Km Athens-Sounio, Anavyssos Attikis 19013, Greece*
[b] *National Centre for Scientific Research "Demokritos", Ag. Paraskevi 153 10, Athens, Greece*

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Abstract

Mussels are worldwide recognized as pollution bioindicators and used in Mussel Watch programs, because they accumulate pollutants in their tissues at elevated levels in relation to pollutant biological availability in the marine environment. The present study deals with the use of *Mytilus galloprovincialis* as a local bioindicator of heavy metal and ^{137}Cs contamination in an estuarine ecosystem (Thermaikos gulf, Greece in Eastern Mediterranean). *M. galloprovincialis* samples were collected monthly from two aquaculture farms during the period April to October 2000. Analyses for the heavy metals Cu, Cr, Ni, Zn, Fe, Mn and ^{137}Cs showed that the concentrations measured were low and similar to those from other non-polluted Mediterranean areas. In terms of the two sampling stations, there were no statistically significant differences between them. On the contrary, the seasonal evolution of either heavy metals or ^{137}Cs levels presented high variation. The levels were found to increase during the cold period of the year, especially for Cu, Zn, Mn and Cr which are essential for life. Stable metals were positively inter-related and moreover, metals more involved in biochemical

* Corresponding author. Tel.: +302102291076371; fax: +302102291076347.
E-mail address: cats@ath.hcmr.gr (V.-A. Catsiki).

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activities seem to present more correlations than others with less significant role in the metabolism of the organisms.

Keywords: Heavy metals; Radionuclides; Caesium; Mussels; Bioindicators; Accumulation; North Aegean Sea

1. Introduction

The trend of marine pollution levels to increase worldwide requires control strategies and routine monitoring of contaminants in the marine environment. Heavy metal and radioactive pollution can cause long-term effects on ecosystems even if their impact has no visible influence in comparison to other pollutants (e.g. effects on organisms can be traced to molecular and cellular responses, as pollution impact does not necessarily lead to observable effects on population or ecosystem). Evaluation of marine pollution levels is based not only on direct measurements of the abiotic components, but also on measurements of the abundance and bioaccumulation of metals and radionuclides in selected marine organisms. The bioaccumulation of contaminants by the tissues and organs of marine organisms has been extensively studied throughout the world and led to the adoption of the bioindicator concept for the environmental quality assessment (Langston and Spence, 1995). Mussels are recognized as pollution bioindicator organisms because they accumulate pollutants in their tissues at elevated levels in relation to pollutant biological availability in the marine environment (Phillips, 1976). Moreover, this ability has led to the adoption of the international ‘‘Mussel Watch’’ program under the CIESM frame and IAEA collaboration and several national programs on Mussel Watch in the marine environment have been carried out (Goldberg et al., 1983; Jernelov, 1996; CIESM, 2002; Claisse, 1989).

The present study deals with the use of *Mytilus galloprovincialis* as a bioindicator of heavy metals and ^{137}Cs contamination in an estuarine ecosystem (Thermaikos gulf, North Aegean Sea, Greece – Fig. 1). The Thermaikos gulf, a complex and interesting ecosystem, is influenced by anthropogenic activities (sewage and industrial discharges, harbor activities) and is the receptacle of three rivers. Axios, Loudias and Aliakmon flowing into Thermaikos gulf after carrying drainage of 93 500 ha of agricultural land. Besides Axios is a trans-boundary river flowing through industrial areas. Previous environmental impact studies have shown elevated values of heavy metals in sediments of Thermaikos gulf, in the vicinity of the pollution sources, namely the industrial zone, the port of Thessaloniki and the sewage outfall (Voutsinou-Taliadouri et al., 1998). The coasts of Thermaikos gulf and its adjacent area host the most extended and productive mussel aquaculture of Greece (70% of the whole Greek production, according to the data of the Greek Ministry of Agriculture (2002)).

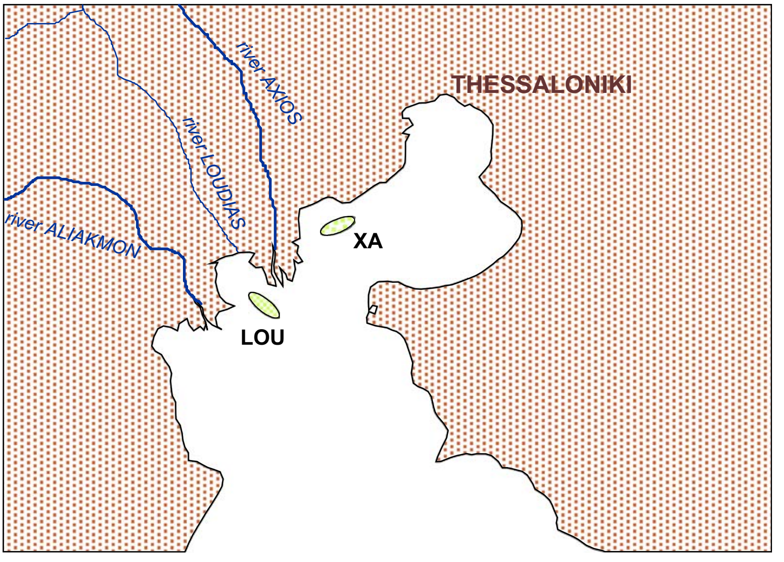

Fig. 1. Location of sampling stations.

The scope of the present study is to elucidate the seasonal behavior of metals (Cu, Cr, Ni, Zn, Fe, Mn and ^{137}Cs) in mussels from an estuarine ecosystem, to provide information on the interrelation of metals, as well as to confirm the use of cultivated *M. galloprovincialis* as bioindicator for heavy metals and ^{137}Cs pollution.

2. Methodology

2.1. Sampling

A total number of 94 composite samples of *M. galloprovincialis* (82 for metals and 12 for ^{137}Cs) were collected monthly from two aquaculture farms located at the areas of Loudias (station LOU) and Halastra (station XA) of Thermaikos gulf (Fig. 1) during the period April to October 2000. The sampling station LOU is located in the wide area of the outflows of the rivers Axios and Loudias and consequently receives agricultural, domestic and industrial discharges carried on by the rivers. The second sampling station (XA) is located to the north, between the coastal area of the city of Thessaloniki and the estuary of the river Axios (Fig. 1). This station is also influenced by the mentioned rivers due to currents – mostly anticyclonic (Papadopoulos, 1996) – as well as, by the domestic discharges and harbor activities of Thessaloniki.

2.2. Heavy metal analyses

A total of 1640 mussel specimens were examined. During each sampling occasion, five to six replicates of composite samples consisting of the soft parts of 20 specimens of similar size were prepared. During each month the size of the specimens was similar, but differed inter-monthly, with the length varying from 3.5 to 7.0 cm. After freeze-drying with a CHRIST GAMMA 1-20 lyophilisator, about 1 g of dried tissue was digested with 10 ml of HNO_3 in a microwave-device (CEM MDS 2100). The dry weight was 22–25% of the fresh weight. After digestion, the sample was diluted with distilled water to 20 ml. A Varian Spectr AA 20 Plus flame Atomic Absorption Spectrophotometer was used for the determination of Cu, Cr, Ni, Zn, Fe and Mn concentrations. The accuracy and precision of the analytical methodology was tested with the reference material NRCC-Dorm-2 of homogenized Dogfish muscle (Table 1).

2.3. ^{137}Cs analyses

Composite samples of at least 80–200 specimens (total weight of about 8–10 kg for each sample) were prepared by adding the soft parts. During each month, the size of the specimens was similar, but differed inter-monthly, with the length varying from 2.3 to 8.5 cm. The samples were ashed using the "dry ashing procedure" for caesium and natural radionuclide measurement (IAEA, 1970). The "ashing factor" was 7.1–14.6% of the fresh weight (defined as the percentage yield of the ash to fresh weight of sample tissue). A quantity of 15–45 g of the ash-sample was put into the appropriate cup ($d = 7$ cm and $h = 1.5$ cm) for gamma-spectroscopy measurement. The samples were measured for 70 000 s in a high-resolution gamma spectrometry system, with an HPGe detector of 20% relative efficiency and computerised multi-channel analyzer of 4000 ch (in a total spectrum area of 2000 keV). ORTEC software was used for the analyses of the obtained spectra. The relative statistical error (1σ) did not exceed 18%. The accuracy and precision of the analytical methodology has been tested by participating in the IAEA's intercomparison exercises for radionuclides in marine reference materials (e.g. during 1993, 1994, 2002).

2.4. Statistical treatment

The statistical treatment of the data, including summary statistics, two-way ANOVA and correlation analysis was performed using the STATGRAPHICS software package.

Table 1
Testing of the analytical methodology with the reference material NRCC-Dorm-2

Metal	Value found	Certified value
Cr (mg/Kg)	36.2 ± 1.6	34.7 ± 5.5
Cu (mg/Kg)	2.17 ± 0.04	2.34 ± 0.16
Ni (mg/Kg)	20.1	19.4 ± 3.1
Zn (mg/Kg)	23.1 ± 0.3	25.6 ± 2.3
Fe (mg/Kg)	142 ± 1	142 ± 10
Mn (mg/Kg)	3.37 ± 0.38	3.66 ± 0.34

3. Results and discussion

The results of AAS and gamma-spectroscopy measurements in mussels from Thermaikos gulf are presented in Table 2 as Bq/kg wet weight for Cs and in μg/g dry weight for the metals. The levels of the measured metals, both radioactive and stable, are characterized in general as low and comparable to those from non-polluted Mediterranean areas.

Considering the heavy metals, the analysed samples showed lower concentrations of Cu and Cr and similar or lower concentrations of Zn, Fe and Mn when compared to the respective ones from Italy (Capelli et al., 1978; Majori et al., 1991; Table 3). Mussels from Turkey had similar levels of Cu, Cr, Fe and Mn and higher levels of Zn and Ni (Kőklű et al., 2000; Topcuoğlu et al., 2002; Table 3).

Statistical analyses (ANOVA) showed that the levels of Cu, Cr, Ni and Zn were generally statistically lower when compared to those from other Greek areas such as Saronikos and Amvrakikos gulfs and Larymna Bay ($P < 0.05$, Fig. 2, Catsiki, 2005). In fact Saronikos presented high levels of Cu and Zn due to sewage wastes, whereas Amvrakikos is a closed gulf having limited communication with the open sea and Larymna Bay is the receptacle of the industrial by-products of a ferro-nickel factory enriched in Cr, Ni and Fe. In contrast, the levels of Fe and Mn measured in mussels from Thermaikos gulf were found to be higher when compared to those from Saronikos and Amvrakikos gulfs (Catsiki et al., 2000; Bei et al., 1998). The mussels sampled from Larymna Bay presented the highest levels of Fe due to the mineral deposits (Florou, 1987; Kozanoglou and Catsiki, 1997).

Considering ^{137}Cs levels, the time evolution of this contaminant is preceding to the present data interpretation, as its presence in the Greek marine environment is mostly related to time determined events (worldwide fallout, impact of the Chernobyl radioactive cloud). The evaluation of ^{137}Cs activity concentrations in seawater and biota in the Aegean and Ionian Sea has been cited by Florou and Kritidis (1994) and Florou (1996). During the early period after the Chernobyl accident, the levels of ^{137}Cs in seawater increased up to one order of magnitude for the Aegean Sea and about 5 times for the Ionian Sea. The late impact of the Chernobyl influence is focused in the North Aegean Sea, where the Black Sea waters interact with the Aegean Sea waters and are diluted. Additionally, the concentrations of ^{137}Cs in sediments and organisms showed an increase up to one order of magnitude higher due to the Chernobyl accident,

Table 2
Summary statistics of contaminants in mussels from Thermaikos gulf (in mg/kg dry weight for the metals and in Bq/kg wet weight for ^{137}Cs)

	Cu	Cr	Ni	Zn	Fe	Mn	^{137}Cs
N	82	82	82	82	82	82	12
Average	3.91	1.85	4.34	56.3	395	14.1	1.23
STD	1.29	0.77	1.39	27.4	159	4.7	0.48
Median	3.55	1.73	4.58	59.6	403	12.7	1.20
Min	1.67	0.20	1.16	20.8	81	7.2	0.3
Max	6.35	3.46	7.51	115.0	715	25.3	1.90

Table 3
Concentration of metals in mussels from Mediterranean and Black Sea

Area	Cu	Cr	Ni	Zn	Fe	Mn		References
Bulgaria	0.5–10.6			12.8–42.0			dw	Stamov and Zlatanova, 1988
Portugal	6.2–13.4			140–542	305–1175		dw	Coimbra et al., 1991
N. Italy	6.9–33.7	5.2–31.6	1.3–11	139–381		9.8–37.8	dw	Capelli et al., 1978
Italy	0.53–5.96	0.18–6.52		10.8–59	11.7–180	1.1–14.2	wet	Majori et al., 1991
Spain	1.1–3			17.3–75.8			ww	Rodriguez et al., 1995
Spain		0.37					wet	Hernandez et al., 1986
Bosporus (Turkey)	2.97–5.65	1.80–2.01		31.9–45.4			dw	Kőklű et al., 2000
Black Sea (Turkey)	7.21–9.1	<0.06–7.58	4.02–24.07	78–512	151–598	5.7–22.8	dw	Topcuoğlu et al., 2002
UK	15.1–40.9	0.6–5.7	5.0–42.9	110–368	215–1072	8.6–27.0	dw	Giusti et al., 1999

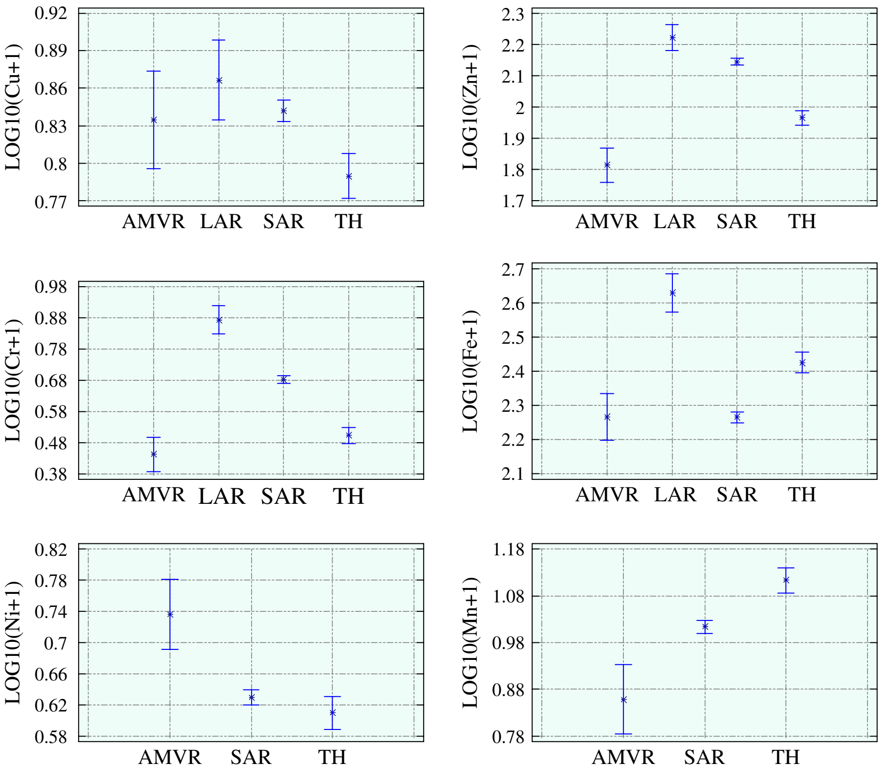

Fig. 2. Mean values and LSD confidence intervals of metals in mussels from diverse Greek areas (AMVR: Amvrakikos gulf, LAR: Larymna bay, SAR: Saronikos gulf, TH: Thermaikos gulf-current data from both sampling stations) (Catsiki, 2005).

whereas since 1987–1988 the ^{137}Cs levels measured in sediments and organisms generally decreased back to the pre-Chernobyl levels. The evolution of ^{137}Cs levels in *M. galloprovincialis* is shown in Table 4.

It is notable that higher levels of ^{137}Cs were detected in the Aegean and Ionian Sea, when compared to the West Mediterranean respective values for 1993: 0.17 ± 0.08 Bq/kg ww in Liguro-Provencial Basin and 0.26 ± 0.06 Bq/kg ww in Alboran Sea (Aarkrog et al., 1994).

In addition, the levels of ^{137}Cs in *M. galloprovincialis* from Amvakikos gulf for the studied period (1.40 ± 0.29 Bq/kg ww) are slightly higher if compared to those reported for Italian (Delfanti et al., 2002) and Adriatic coasts as well (Kniewald and Barisic, 2002).

Concerning the two sampling stations, the respective mussel samples did not show, in general, statistically significant differences in the observed levels of heavy metals and ^{137}Cs ($P > 0.05$). However, the samples from Loudias area (LOU) tended to have higher mean values compared to those from Halastra (XA) for the most metals (Cr, Cu, Zn, Mn and ^{137}Cs). This is probably due to the location of the LOU station among the

Table 4
^{137}Cs activity concentrations (Bq/kg wet weight) in *Mytilus galloprovincialis* in the Eastern Mediterranean – Aegean and Ionian Sea[a], (Florou et al., 2002)

Time period (y)	^{137}Cs (Bq/kg wet weight)	Remarks
1984–85	0.68 ± 0.10	
1986–87	7.10 ± 1.80	Excluded maximum observed 50 ± 7 (3 samples from one station)
1988–95	0.46 ± 0.28	
1999–2001[b]	0.92 ± 0.68	Mean value ± Standard deviation of (present study)
LLD	0.1	Measurement in dried soft tissue transformed into wet weight

[a] The respective values from Ionian Sea vary in the same range (sparse samples).
[b] The limited data of 2002 are in the same range as for 2001.

three rivers outflow (Aliakmon, Loudias, Axios – Fig. 1) and to anticyclonic currents (Papadopoulos, 1996), which enforce the pollutant persistence in the area.

The time series values of heavy metals (average of five to six replicates) and ^{137}Cs (activity concentrations of composite samples) are presented in Fig. 3. The seasonal evolution of heavy metal and ^{137}Cs levels during the study period presented high variation. It can be seen that two groups of temporal patterns are followed: one by Cs–Ni–Fe with minima during spring (April) and autumn (October) and one by Cu–Zn and Mn–Cr with maxima during the above seasons.

In order to clarify the above seasonal pattern in mussels from the estuarine ecosystem of Thermaikos, the metal data were grouped in three periods named as: "a" comprising samples collected during April (early spring), "b" comprising samples collected during May to July (summer) and "c" comprising samples collected during August to October (early autumn). These data were treated statistically by a two-way ANOVA to determine the seasonal differences in the two sampling stations. "Station" and "season" were the categorical factors. Because mussel size influences contaminant accumulation, the shell length was used as a covariate parameter.

Based on the results of the two-way ANOVA (Fig. 4), some comments can be made as follows. The seasonal patterns for the metals Cu, Zn, Mn and Cr are more or less similar ($P < 0.001$), with the minimum values during the summer (season "b") and maximum values mostly during the early spring (season "a"). In contrast, the second metal group of Ni, Fe and ^{137}Cs showed the maximum values during the summer (season "b") only in the station LOU ($P < 0.001$). Considering the station XA (Halastra), the seasonal evolution of Ni showed the same pattern to the station LOU (Loudias), whereas the pattern followed by Fe and ^{137}Cs showed the minima during summer (Fig. 4).

For comparison reasons, data from Saronikos gulf (Greece) were evaluated for the years 1999 and 2001 (Fig. 5). These data were obtained in the framework of the Greek MED.POL Program (Catsiki et al., 2001). Statistical treatment of these data revealed that all the studied metals (Cu, Zn, Mn, Cr, Ni, and Fe) have the same seasonal evolution pattern with the minimum values observed during the summer

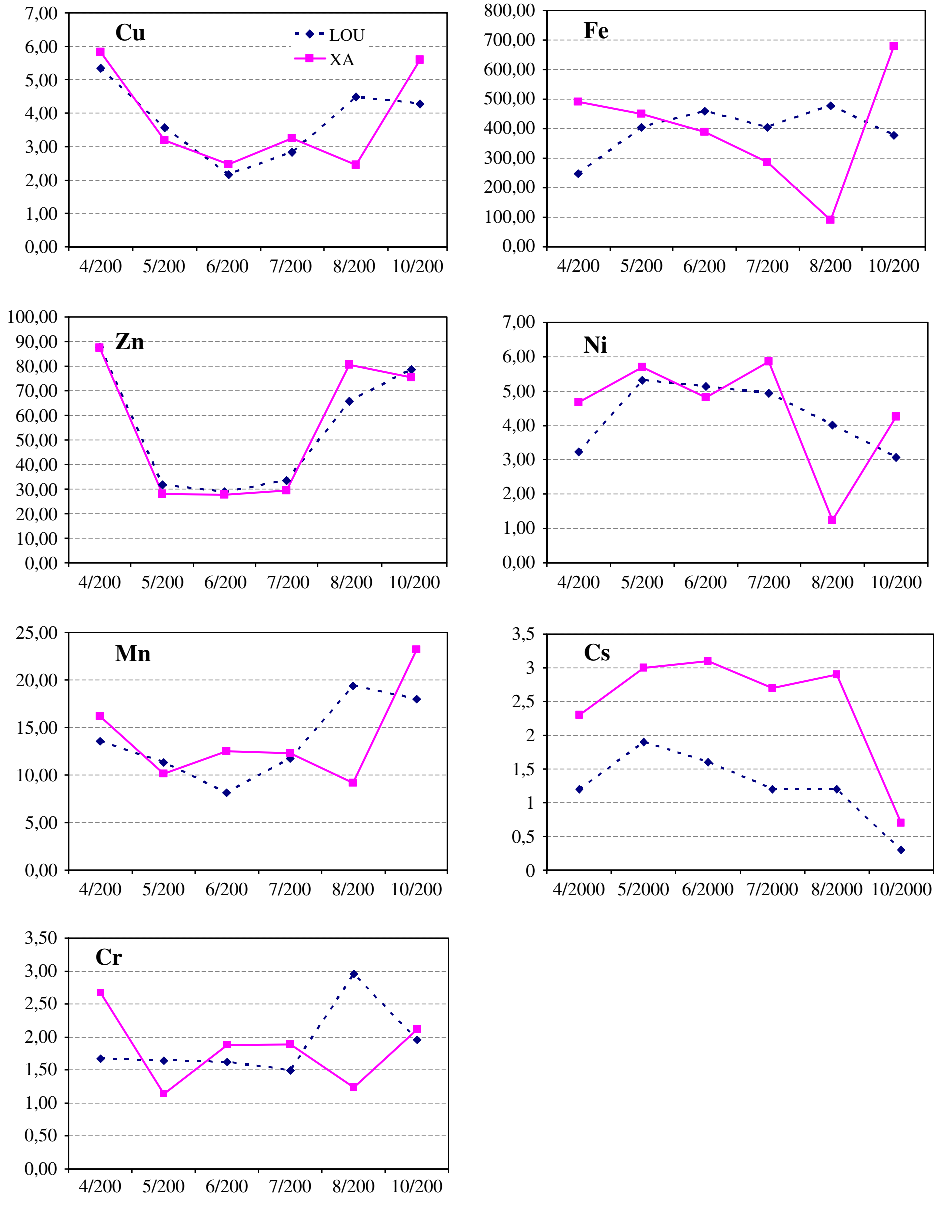

Fig. 3. Temporal evolution of metals (average of five to six replicates per sampling) and ^{137}Cs activity (concentrations of composite samples) in mussels. Concentrations in μg/g dw for metals and in Bq/kg ww for ^{137}Cs.

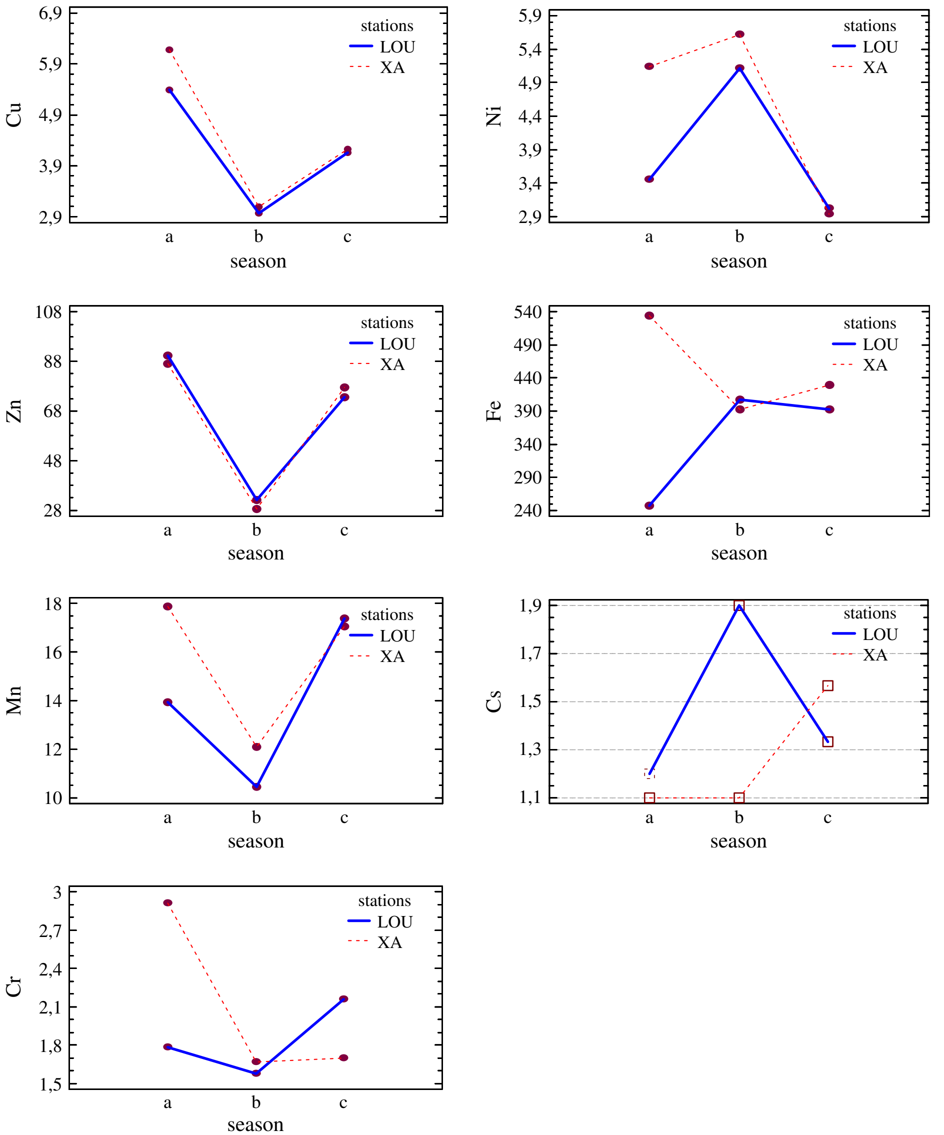

Fig. 4. Seasonal evolution of metals and ^{137}Cs in mussels of Thermaikos gulf. Concentrations in μg/g dw for metals and in Bq/kg ww for ^{137}Cs. Season "a" = April (spring), "b" = May to July (summer) and "c" = August to October (autumn).

(season "b") – fact also observed in Thermaikos gulf (group of Cu, Zn, Mn, Cr). It is worthy to note that during the cold period, the examined mussels were found to be at the life stage of spawning (more or less mature gonads).

It is known that the bioaccumulation of metals by mussels varies according to the season. Seasonal bioaccumulation patterns indicate the effect of several environmental

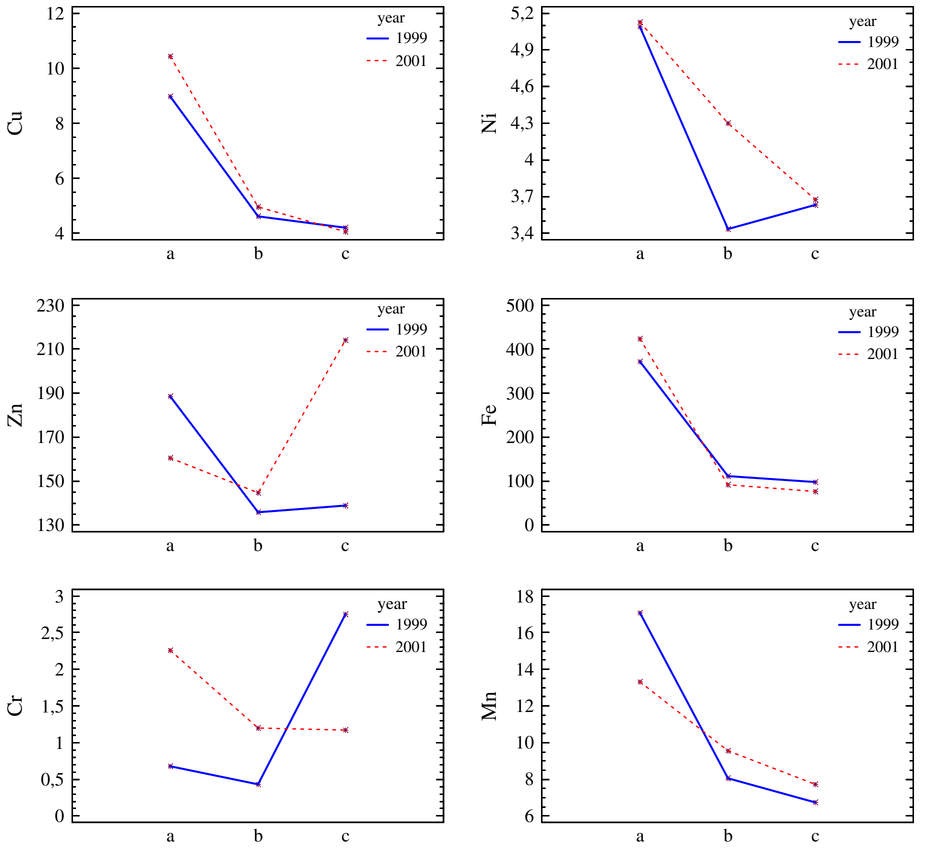

Fig. 5. Seasonal evolution of metals in mussels of Saronikos gulf (in μg/g dw, "a" = February to April (spring), "b" = May to July (summer) and "c" = August to October (autumn). (Catsiki et al., 2001).

and biological factors acting individually or associated, competitively or synergistically. In the studied case, one can assume that mussels during the warm period bioaccumulate in a lesser degree than during the colder period. This finding can be attributed either or both to biological parameters e.g. spawning time or/and environmental ones, such as river outflow, snow melting etc. As it has been reported, the reproductive cycle of mussels influence their metal content, increasing it during the period before spawning (Cossa, 1989; Bryan, 1976; Catsiki and Strogyloudi, 1999).

Nevertheless, differences among the temporal evolution patterns of metals as well as differences between levels in different sampling localities, are often inter-related and consequently smoothed or/and masked. Covariance analyses is a useful tool for studying whether metals are bio-accumulated all with a common rate or their assimilation is directed by specific rules. The covariance analyses using the obtained data showed that the bioconcentrated metals were positively inter-correlated ($P < 0.05$ and $P = 0.09$ – Fig. 5). This means that the studied metals increased or decreased simultaneously. It is relevant that Cu, Fe, Zn and Mn are essential for life in reasonable amounts and required by biota for various biochemical processes. Chromium is less abundant and Cs does not seem to have any known metabolic role in the physiology of organisms. Moreover, as it has been reported (Catsiki and Strogyloudi, 2000), metals such as Fe, Zn and Cu, which are involved in many biochemical activities, seem to present more correlations than others with less significant role in the metabolism of the organisms.

One can note in general that the biological parameters play the main role in the bioaccumulation of the essential elements, whereas for those with no metabolic role the environmental parameters are the affecting factors.

4. Conclusions

The accumulation of stable (Cu, Cr, Ni, Zn, Fe, Mn) and radioactive metals (^{137}Cs) by the mussels of the estuarine ecosystem of Thermaikos gulf presented high seasonal variability, strongly influenced by the abiotic (environmental) parameters of the marine environment and the biological ones (reproductive cycle of mussels). The levels were found to increase during the cold period of the year, especially for Cu, Zn, Mn and Cr which are essential for life. Stable metals were positively inter-related and moreover, metals more involved in biochemical activities seem to present more correlations than others with less significant role in the metabolism of the organisms.

The levels of the measured metals from North Aegean Sea (Eastern Mediterranean), both radioactive and stable, are characterized in general as low and comparable to those from other non-polluted Mediterranean areas.

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