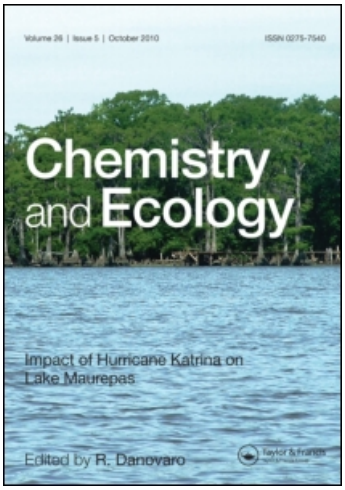

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Trace metal pollution in surface marine waters: Nearshore concentrations along Apulia and Albania

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TRACE METAL POLLUTION IN SURFACE MARINE WATERS: NEARSHORE CONCENTRATIONS ALONG APULIA AND ALBANIA

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The concentrations of some heavy metals (Zn, Cu, Mn, Pb, Ni and Cd) were measured in surface coastal waters of the southern Adriatic Sea, in the framework of the Interregional Program II (INTERREG Italy–Albania). Seven stations were positioned along the Albanian shoreline, at a distance of 500 m from the coast, while the remaining stations were located on inshore–offshore transects along the Adriatic side of Apulia. Concentrations exhibited relatively low values, lower (or similar) than those observed in other Italian coastal areas and generally much lower than at other sites of the coastal Mediterranean. Lead (and to a lesser extent cadmium) showed the most even distribution throughout the study area, most likely resulting from atmospheric inputs. Harbours and river outlets were identified as critical points for the input of the remaining trace elements along both Apulia and Albania. In the large majority of stations, bioassays with the sea urchin *Paracentrotus lividus* highlighted moderate toxic effects of sea water on these organisms. Heavy metal concentration levels measured in the study area are not directly related to embryotoxicity, probably because they are not high enough to determine toxic effects upon biota, and/or because elements other than metals are of major importance in the coastal area of the southern Adriatic Sea.

Keywords: Sea water; Heavy metals; Southern Adriatic sea

1 INTRODUCTION

Over the past two decades considerable attention has been given to problems dealing with the adverse effects of heavy metals on various ecosystems and several studies have been conducted in the Mediterranean Sea, aimed at determining their concentrations and distribution in different environmental media. Heavy metals neither biodegrade nor can be eliminated by incineration processes. They tend to be persistent pollutants and can accumulate in both abiotic compartments (soils, freshwater, sea water and sediments) and biota. Each metal has a specific chemical form (speciation) which determines its solubility in sea water and consequently its ability to incorporate into biological systems (Salomons and Forstner, 1984). In the marine environment, trace concentrations of these elements can negatively

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affect the marine biota and pose risk to human consumers of seafood (Bryan, 1976, 1984; Langston, 1990).

Although trace metals are natural constituents of the Earth's crust and are present in all ecosystems, their concentrations have been dramatically increased by human activities (e.g. Guerzoni *et al.*, 1984; Dall'Aglio and Fornaseri, 1988; Correggiari *et al.*, 1989; Ridame *et al.*, 1999). Natural sources of heavy metals include wind blowing of dust from the soil surface, volcanic activity, and formation of sea salt particles. Natural emissions due to volcanic activity are intense in southern Italy, due to the presence of several active volcanoes (http://www.msceast.org/hms/emission.html). The main anthropogenic sources are industrial activities, including present and past mining activities, foundries and smelters. Diffuse sources (e.g. piping, combustion by-products, transports) may also be important, and favour the widespread diffusion of these contaminants in the environment. For example, relatively volatile compounds and those that become attached to airborne particles can be widely dispersed on very large scale, conveyed to sea water and sediments and be largely retained within the coastal and shelf regions. The overall anthropogenic impact upon the coastal region is a mosaic composed by the different types of inputs and mechanisms, some of which are predictable whereas some others might not be. Besides heavy metals, land-based sources of pollution may introduce a large variety of pollutants into the marine environment, including persistent organic pollutants such as polychlorinated biphenyls, organochlorine pesticides, dioxins and furans, organophosphorous compounds, polycyclic aromatic hydrocarbons.

Although it is undoubtedly important to assess the levels of chemical contamination of natural systems, the analytical determination of contaminants can hardly address all possible types of substances present in the environment and moreover it does not provide any information about the possible impacts upon the biota, which may depend on factors other than the amount and type of xenobiotics present in the system (e.g. their bioavailability). Living systems integrate the complexity of effects exerted by biologically active contaminants, which never produce a simple addition of effects but are largely synergetic (Stebbing, 1985). Therefore, for the assessment of ecosystem health, it is worth considering both chemical parameters in the ambient matrices (water or sediments) and their impact on the biota (Chapman and Long, 1983). This latter aspect is usually achieved by conducting experimental bioassays, where the biotic responses by well known 'test species' are considered as indicators of the quality of the medium to which organisms have been exposed and, finally, to the potential toxicity of their habitat (Chapman and Long, 1983). Sea urchins are among the most commonly used organisms in this type of bioassay. The choice of sea urchins for this kind of assessment offers several advantages, including their large occurrence in coastal waters, the possibility to perform bioassays on several developmental stages (embryos, gametes or adults) and to observe different biological events (e.g. reproductive success, embryogenesis, mitotic activity and bioaccumulation) (Pagano *et al.*, 1986).

2 MATERIALS AND METHODS

In the framework of the Interregional Program II (INTERREG Italy–Albania), marine superficial waters were collected from April to May 2001 at 29 stations in the southern Adriatic Sea, for the determination of heavy metal concentrations. Seven stations were positioned along the Albanian shoreline, at a distance of 500 m from the coast, while the remaining stations were located on inshore–offshore transects along the Adriatic side of Apulia (Tab. I).

TABLE I Sampling Stations in the Southern Adriatic Sea.

No. of Stations	*Location*	*Longitude*	*Latitude*	*Depth (m)*
Albania				
2	River Drinit	19° 30.81′	41° 48.88′	36
4	Lezhe	19° 32.00′	41° 41.05′	26
6	River Ishmit	19° 23.51′	41° 29.95′	23
9	Durrës	19° 24.00′	41° 02.59′	26
14	River Semanit	19° 18.40′	40° 47.50′	18
16	Valona	19° 18.50′	40° 21.98′	113
17	–	19° 33.86′	40° 05.76′	255
Apulia				
23	Otranto	18° 43.22′	40° 07.10′	363
24	Otranto	18° 32.06′	40° 05.46′	87
25	Lecce	18° 19.06′	40° 25.12′	71
26	Brindisi	18° 07.26′	40° 40.81′	96
27	Brindisi	18° 15.23′	40° 45.03′	128
28	Brindisi	18° 28.09′	40° 50.62′	766
29	–	17° 41.51′	40° 47.54′	58
30	–	17° 55.48′	40° 55.48′	123
33	Bari	17° 05.03′	41° 05.03′	134
34	Bari	16° 55.44′	41° 55.44′	69
35	G. Manfredonia	16° 33.11′	41° 33.11′	37
36	G. Manfredonia	16° 52.99′	41° 52.99′	123
37	G. Manfredonia	17° 04.48′	41° 4.48′	392
38	G. Manfredonia	17° 01.71′	41° 46.38′	711
39	G. Manfredonia	16° 49.13′	41° 42.64′	139
40	G. Manfredonia	16° 36.24′	41° 37.08′	86
41	G. Manfredonia	16° 28.20′	41° 26.06′	42
42	G. Manfredonia	16° 13.23′	41° 27.04′	19
43	G. Manfredonia	16° 14.21′	41° 42.83′	12
44	G. Manfredonia	16° 02.50′	41° 33.50′	12
45	G. Manfredonia	16° 40.98′	41° 49.33′	109
46	G. Manfredonia	16° 54.91′	41° 53.52′	440

To avoid contamination from the ship, water samples were collected a few meters below the sea surface and immediately filtered through 0.45-μm pore-size Millipore filters, to remove particulate matter. Filters were previously acidified with a 10% nitric acid solution. After filtration, water samples were acidified with 0.5% nitric acid, to hinder the deposition of heavy metals on the inner surface of container, stored at dark and at 4 °C. Trace metals were analyzed by electrothermal vaporization inductively coupled plasma mass spectrometry, following the procedure of Chapple and Byrne (1996) for Mn, Ni and Cu, and Rosland and Lund (1998) for Pb, Cd and Zn.

In addition to heavy metal determination, water toxicity was evaluated through embryotoxicity tests conducted on the sea urchin *Paracentrotus lividus.* Embryos of this species were exposed to the collected sea water during 48 h and toxicity was deduced from the percentage of anomalous plutei, i.e. individuals characterised by skeletal malformations, gut abnormalities or inhibition of larval development resulting in a size $<1/2$ that of normal larvae. To calculate toxicity, anomalous plutei were considered all together, without distinction among the different abnormalities (Pagano *et al.*, 1986). Toxicity was expressed as percentage effect: percentage effect $= ((N_c - N_s)/N_c) \times 100$, where N_c and N_s are the average normal plutei numbers in the control medium and water sample, respectively. The control medium was filtered sea water, collected in the same area where the urchins were sampled. It was considered suitable for the calculation of percentage effect when normal plutei were $\geq$80% of total larvae.

3 RESULTS AND DISCUSSION

In the whole study area average concentrations of the analysed trace metals equaled (in decreasing order) 4.74 ppb for zinc, 1.77 ppb for copper, 1.28 ppb for manganese, 0.58 ppb for lead, 0.44 ppb for nickel and 0.04 ppb for cadmium. Table II reports the range of values and (when possible) mean concentrations of heavy metals in our study area and other Mediterranean waters. Background reference concentrations (BRC) are also reported (OSPAR, 2000), in order to give an idea of the concentrations that would be expected to be found in sea water in the absence of any human activity. These concentrations are rather similar to the values measured in Mediterranean open waters, but of course they are much lower (up to three orders of magnitude for lead) than the concentrations measured in our study.

Except zinc, the analysed elements showed higher concentrations along Albania. Although ranges of values were similar along Albania and Apulia, the spatially integrated means were double (Mn and Ni) to 6–7 times higher (Cd and Pb) in Albanian coastal waters (Tab. II).

The concentrations of most trace metals in the southern Adriatic Sea were generally low and showed lower (or similar) values as compared to other Italian coastal areas and much lower values than in other Mediterranean coastal waters. Cadmium average concentration in the whole investigated area (0.04 ppb), for example, was one magnitude lower than near the Eolian Islands (Dall'Aglio and Fornaseri, 1988) or along the Campania shore (Manfra and Accornero, 2002). The same applies to zinc (4.74 ppb), which showed an average concentration of about one-half than in Campania nearshore waters (Manfra and Accornero, 2002). Maximum concentrations measured for nickel and copper were similar to maximal values found near the Italian shoreline but 4–10 (Cu) and 30 (Ni) times lower than in other sites of the coastal Mediterranean. Conversely, lead was the only element that exhibited higher concentrations in the Adriatic than on the Tyrrhenian side.

Among all heavy metals, lead (and to a lesser extent cadmium) showed the most even distribution throughout the study area and a relatively restricted range of concentration variability (Fig. 1).

TABLE II Range of values and mean concentrations (in parentheses) of heavy metals (μg/l) in sea water.

Location	*Cu*	*Pb*	*Cd*	*Zn*	*Ni*	*Mn*
[a]BRC (OSPAR area)	0.05–0.36	0.005–0.02	0.005–0.02	n.d.	n.d.	n.d.
[b]Open Mediterranean Sea	0.04–0.70	0.018–0.14	0.004–0.06	0.24–0.56	<0.5[c]	n.d.
[d]Southern Adriatic Sea (Albania)	0.30–4.87 (2.32)	0.04–6.53 (1.63)	0.01–0.22 (0.11)	0.07–14.09 (4.13)	0.11–1.36 (0.75)	0.04–4.82 (2.06)
[d]Southern Adriatic Sea (Apulia)	0.50–5.00 (1.60)	0.04–2.93 (0.24)	0.01–0.25 (0.02)	0.07–20.94 (4.94)	0.11–1.26 (0.34)	0.04–4.52 (1.03)
[e]Campania coastal waters	0.60–6.74 (2.58)	0.06–2.51 (0.25)	0.01–1.30 (0.12)	0.07–69.7 (8.04)	0.09–1.0 (0.35)	0.04–20.46 (1.65)
[f]Eolian islands	0.04–3.20	0.15–0.62	0.18–0.83	n.d.	n.d.	n.d.
[g]Greek coastal waters	0.03–20.7	0.03–12.2	0.002–2.3	0.02–120	0.06–41.9	0.02–13.0
[b]Coastal Mediterranean Sea	0.01–50	0.016–20.5	0.002–0.9	0.20–210	n.d.	n.d.

Note: n.d., not determined.
[a]BRC background reference concentrations (OSPAR, 2000).
[b]UNEP (1996).
[c]UNEP (1993).
[d]This study.
[e]Manfra and Accornero (2002).
[f]Dall'Aglio and Fornaseri (1988).
[g]Dassenakis *et al.* (1996).

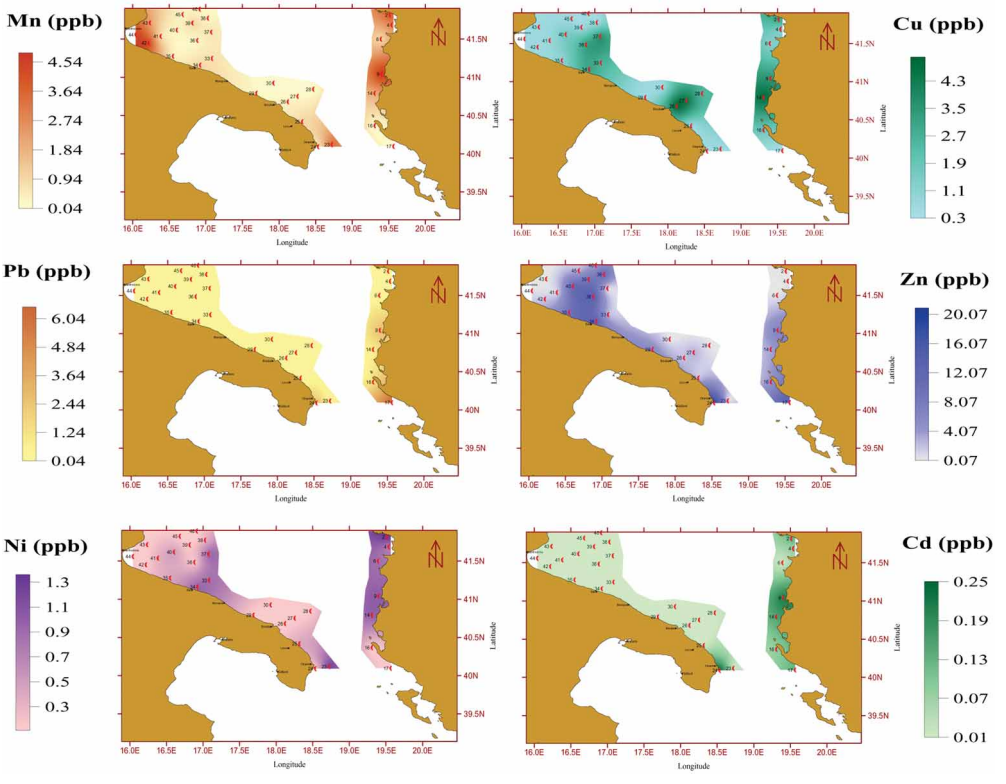

FIGURE 1 Sea water heavy metal concentrations in the southern Adriatic Sea.

This is most likely because the major source of these elements in the region originates from atmospheric inputs (e.g. Elbaz-Poulichet *et al.*, 2001). On the basis of comparisons between aeolian dust and marine samples, Correggiari *et al.* (1989) found that riverine and terrestrial inputs represented negligible sources of Pb and Cd as compared to atmospheric deposition in the central Mediterranean (Tyrrhenian and Adriatic Seas), despite the general diminution of their atmospheric fallout resulting from the reduction of emissions (Ridame *et al.*, 1999). In particular, sea water cadmium is highly homogeneous, both spatially and vertically, over the whole western Mediterranean basin, as a result of the combined effects of atmospheric deposition, which prevents surface depletion, and active vertical mixing, which attenuates surface enrichment (Copin-Montégut *et al.*, 1986).

On the contrary, trace elements such as copper and zinc (and manganese and nickel on the Albanian side) showed a clear trend in distribution. In the whole study area harbours and river outlets represent critical points for their input into marine coastal waters (Fig. 1), similarly to other areas in the Mediterranean (Evison and Tosti, 1980).

Trace elements showed relatively low concentrations along the Apulian seashore, with the exception of the transects in front of Brindisi (Sts. 26, 27, 28) and Bari (Sts. 33, 34), where concentrations tended to decrease from the most coastal station offshore (Fig. 1). Cu concentrations halved and Zn concentrations dramatically diminished over a distance of a few tens of kilometres, showing the same onshore–offshore gradient which characterises sediment concentrations (Ianni *et al.*, 2002), hence suggesting that coastal sources (and subsequent dilution with distance from the coast) are the most important factor in determining the

distributions of those elements in these areas. In the Gulf of Manfredonia (Sts. 35–46) sea water metals were generally less abundant than near Brindisi and Bari and sediment concentrations essentially depend on granulometry (Ianni *et al.*, 2002). Although the Manfredonia area belongs to the 15 'sites of national priority' identified by the Italian law (Legge 426/98) for decontamination, terrestrial sources of heavy metals do not seem to severely affect the contiguous marine area. This probably results from the fact that the most impacting industries (Enichem, Snia-Anic) are currently inactive and minor active plants are equipped with industrial waste disposal systems (http://www.investinitaly.com.dd3puglia_manfredonia.htm). In spite of this, soils and underground waters of the same area are being characterized by widespread contamination of substances supplied by past productivity cycles, such as arsenic and mercury, which largely exceed the allowed concentrations (e.g. Hg $\approx 176\,\mathrm{mg\,kg^{-1}}$, versus the allowed limit of $5\,\mathrm{mg\,kg^{-1}}$). Conversely, Brindisi (http://www.porto.brindisi.it/porto/caratt.html; http://www.porto.brindisi.it/stat/stat_2002.html) and, even more, Bari (http://www.porto.bari.it/porto/caratt.html; http://www.porto.bari.it/stat/stat_2002.html), (which are also part of the above-mentioned 'sites of national priority for decontamination', with their polyvalent activities, the big harbour and the related shipping activities, seem to significantly contribute to the degradation of the marine environment, mainly through the input of hydrocarbons and metallic contaminants.

On the Albanian side, the waters adjacent to Durrës (St. 9), the Semanit (St. 14) and, to a lesser extent, the Drinit (St. 2) river mouths exhibited the highest concentrations of trace metals (Fig. 2). Despite the difficulty in accessing to information regarding pollutant sources in Albania, local water pollution from industrial and domestic effluents is known to contribute in affecting Mediterranean sea water quality (Gjika, www.imbc.gr/biblio_serv/medcst/X0137_123.html). Rivers are the vectors of toxic substances from a variety of sources, including domestic, industrial and agricultural effluents, and it is reasonable to hypothesise that river impact is even higher in countries where productive technologies are generally outdated and the critical social situation lead to consider economical enrichment as a priority compared to the respect of the environment. There is virtually no wastewater treatment in

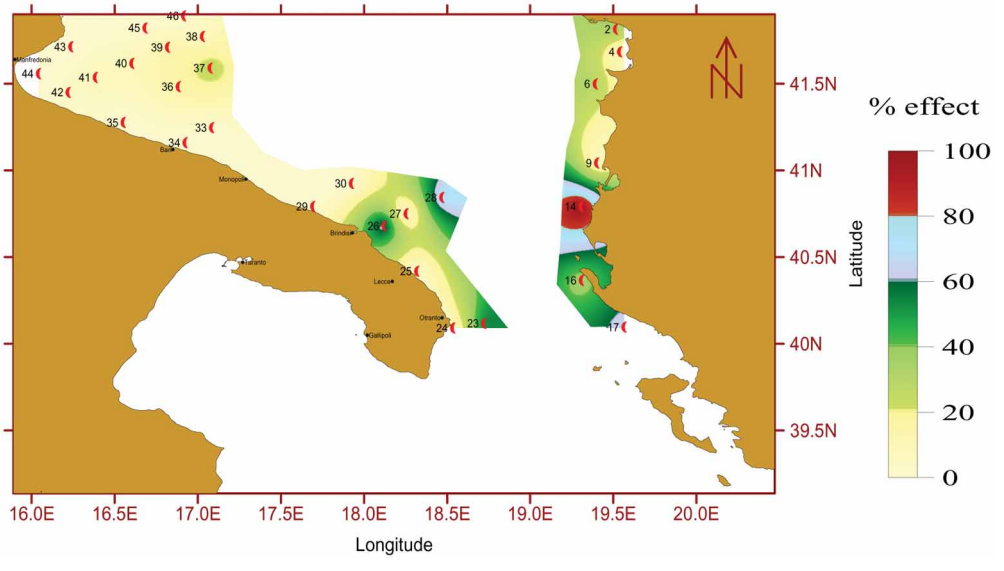

FIGURE 2 Sea water toxicity (tested by *P. lividus*) in the southern Adriatic Sea.

Albania. Communal and industrial wastewaters are discharged directly into receiving waters. Industrial pollution sources include makers of cement, leather, ceramics, textiles, as well as mines, smelters, oil and gas producers, and wood processing facilities (UNEP, 2000). The rivers discharging in the northernmost part of Albania pass through the richest mineralogical area of the country (chromium and copper mainly) and receive the wastewaters of an important copper refinery and of a plant processing iron–chromium alloys. However, the most important source of heavy metals along the Adriatic side of the country is Durrës. Besides being the largest and most important harbour of Albania, Durrës is located in one of the most severely contaminated areas of the country (Baraj *et al.*, 1994).

To our knowledge, this is the first study investigating the concentration of heavy metals in coastal waters of the southern Adriatic Sea. Sea water concentrations are generally difficult to determine, because of the very high concentration of the matrix elements (in particular sodium chloride) and the very low concentration of some of the trace metals. Generally, sea water concentrations of trace elements are at least one order of magnitude lower than concentrations in sediments or benthic organisms at the same site (e.g. Fowler, 1990). Metallic contaminants may accumulate in sediments at concentrations 1000–5000 times higher than in the water column (Livett, 1988). Similarly to water concentrations, metal amounts in sediments (Viel *et al.*, 1986; Storelli *et al.*, 2001; Ianni *et al.*, 2002) and benthos (Storelli *et al.*, 2001) from the investigated area generally show, with a few exceptions, modest levels of contamination. Sediment concentrations along Apulia account for up to 25 ppb Cu, 6.69 ppb Pb, 0.24 ppb Cd, 151.2 ppb Zn, 30 ppb Ni and 496 ppb Mn (Storelli *et al.*, 2001), showing values similar to those measured in sea water for Pb and Cd, and 5 times higher in sediments for Cu, 7 times higher for Zn, 20 times higher for Ni and 100 times higher for Mn. The occurrence of metals in the tissues of green algae, holothurians and sea urchins is also low (Storelli *et al.*, 2001), especially if compared to that reported in other coastal areas impacted by strong anthropic pressure (Haug *et al.*, 1974; Agadi *et al.*, 1978).

Similarly to water concentrations, sediment amounts of heavy metals near Albania are higher than near Apulia, attaining maxima of 51.1 ppb Pb, 1.16 ppb Cd and 1930 ppb Mn in the Durrës-river Ishmit area, and 624 ppb Cu, 355 ppb Zn and 413 ppb Ni in the northern sector of the coastal zone (Çelo *et al.*, 1999).

Independent of the low heavy metal concentrations found in sea water and reported for sediments and the biota, bioassays with the sea urchin *P. lividus* were performed in this study (Manfra, 2001), in order to evaluate the overall toxicity of the samples (see Introduction).

Percentage effect was <40% in all stations north of the river Semanit (St. 14), which indicates low environment toxicity, considering that values <20% are looked at as non-toxic (International Organization for Standardization, 1998). The toxicity of samples was generally not high enough to hinder the development of *P. lividus* embryos, although it caused retards and malformations on the skeleton and gut of larvae. The highest toxic effects were observed along the transect in front of Brindisi (Sts. 26 and 28), in agreement with chemical analyses, and in the southernmost Albanian stations (Sts. 14 and 17). This latter area is the site of a chlor-alkali plant discharging its wastes into the sea, and is the most Hg-contaminated district of the country (Baraj *et al.*, 1994). On the contrary, in other sites coastal waters did not highlight significant levels of toxicity, although heavy metals concentrations were relatively high (e.g. Bari). Since the sea urchin larval bioassay is a suitable test to evaluate the toxicity resulting from the presence of heavy metals (His *et al.*, 1999), the lack of correspondence between chemical and ecotoxicological results probably depends on the fact that concentration levels like those observed in this study, alone, are not high enough to determine toxic effects upon biota, and/or elements other than metals are of major importance in the coastal area of the southern Adriatic Sea.

4 CONCLUSION

In synthesis, the results of this study lead to the following concluding considerations:

1. The occurrence of heavy metals in coastal superficial waters of the southern Adriatic Sea is low, especially when compared with that observed in other Italian or Mediterranean coastal areas.
2. While Pb and Cd showed an even distribution throughout the study area, most likely resulting from widespread atmospheric inputs, critical points for the input of the remaining elements were represented by harbours and river outlets. Brindisi and Bari along the Apulian coast and Durrës in Albania were identified as hot spots of land based heavy metal pollution.
3. The metal concentration levels measured in the study area do not seem to be directly related to the adverse effects observed for some sites upon embryos of sea urchin *P. lividus.*

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