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RESEARCH ARTICLE

Heavy metal accumulation in *Diplodus annularis*, *Liza aurata*, and *Solea vulgaris* relevant to their concentration in water and sediment from the southwestern Mediterranean (coast of Sfax)

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Abstract The concentrations of heavy metals (Cd, Cu, Fe, Pb, Ni, and Zn) were measured in the liver, gills, and muscle of *Solea vulgaris*, *Liza aurata*, and *Diplodus annularis*, collected from the south coast of Sfax (Gabes Gulf, southwestern Mediterranean). The concentrations of heavy metals in water exhibited the following decreasing order (expressed in $\mu g\ l^{-1}$): Fe > Ni > Zn > Cu > Pb > Cd whereas the trend is somewhat different in sediments (mg kg^{-1} D.W.) Fe > Zn > Pb > Ni > Cu > Cd. The levels of heavy metals varied significantly among fish species and tissues. Heavy metal levels were found generally higher in the liver and gills than the muscle in all species. The liver was the target organ for Cd, Cu, Fe, Ni, and Zn accumulation. Nickel and lead, however, exhibited their highest concentrations in the gills. The three studied fishes showed a difference in metals accumulation decreasing in following order *S. vulgaris* > *D. annularis* > *L. aurata*. *Solea vulgaris* with the highest TF_{water}, $TF_{sediment}$, and metal concentrations in tissues would be considered as a potential bio-indicator in the south coast of Sfax for the assessment of environmental pollution status. Comparative studies with Luza zone indicate considerable bioaccumulation of heavy metals (Pb and Zn) in the various tissues of fish samples of the south coast of Sfax.

Keywords Bioaccumulation · Heavy metals · Sfax south coast · *Liza aurata* · *Solea vulgaris* · *Diplodus annularis*

Introduction

Gabes Gulf (southwestern Mediterranean) is considered the main seafood resource contributing about 65 % of the national fish production in Tunisia (D G P A 2004; Ben Rebah et al. 2010). However, due to the increase of urbanization, industry, overfishing, tourism, and the discharge of huge amounts of phosphogypsum and other pollutants, this gulf has been reported to be densely polluted (Hamza-Chaffai et al. 1997). The city of Sfax is one of the main harbors of the Gulf of Gabes and an important industrial center whose pollution level is in contrast with the nearby and beautiful Kerkennah Island (Zaghden et al. 2005). The south coast of Sfax is under a high environmental pollution pressure and concentrates many industrial and anthropogenic activities. Previous studies have shown that this area was mainly affected by heavy metal pollution, mainly related to the industry of phosphates as well as other heavy metal transmitters such as salt works, tanneries, textiles, lead foundries, soap factories, ceramics industries, and building materials (Hamza-Chaffai et al. 1995; Barhoumi et al. 2009; Kessabi et al. 2009; Messaoudi et al. 2009a, b; Smaoui-Damak et al. 2009; Rabaoui et al. 2013; Ben Salem et al. 2015).

Due to their toxicity, persistence bioaccumulation in the environment, and ecological risks, heavy metals are of increasing global concern (Zhou et al. 2007; Gao and Chen 2012; Gu et al. 2012b). Heavy metals are categorized as potentially toxic (e.g., Cd, Pb, and Ni) and essential (e.g., Cu, Zn, and Fe). Even at low concentrations, toxic metals can be very harmful to human health when

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ingested over a long time period. Essential metals can also produce toxic effects with excessive intake (Ashraf et al. 2006; Uluozlu et al. 2007; Tuzen 2009).

In aquatic ecosystems, organisms may be considered as bio-indicators in determining the impact of toxic heavy metals (Arain et al. 2008), an approach that was used in the south coast of Sfax. Since they are at the top of the aquatic food chain, fish can accumulate heavy metals from food, water, and sediments and are widely used to biologically monitor the degree of metal pollution in aquatic ecosystems (Chovanec et al. 2003; Brumbaugh et al. 2005; Yılmaz et al. 2007; Al Sayegh Petkovšek et al. 2012; Zhao et al. 2012). The specific advantages of using fish are the following: (1) they are long living and absorb a variety of pollutants over time, (2) they live in water and/or sediment and enable continuous surveillance of pollutant presence, and (3) they are easily sampled.

The study of metal accumulation in fish tissues (gills, liver, and muscle) is of interest in assessing pollution in sea water. These tissues in fish were chosen because (1) the liver is the main accumulation organ for biotransformation and excretion of pollutants, including metals (Moon et al. 1985; Triebskorn et al. 1997); (2) the gills reflect the metal levels in the water since they are in direct contact and are the primary sites of gas exchange, acid–base regulation, and ion transfer (Randall 1990); and (3) the muscle is the part consumed by humans.

Heavy metals accumulate as they move up the food chain and may reach dangerous levels for human health (Ip et al. 2005; Sapkota et al. 2008; Yi et al. 2011; Gu et al. 2012a). The *Diplodus annularis* (Linnaeus, 1758), benthopelagic species common in the bottoms covered by Posidonia (Derbal et al. 2007; Chaouch et al. 2013); *Liza aurata* (Risso, 1810), pelagic species often encountered in estuaries, backwaters, and inshore areas of the Mediterranean; and *Solea vulgaris* (Quensel, 1806), benthic species feeding on organisms living in the sediments are important target species for Tunisian fisheries and particularly for those in the Gulf of Gabes. Thus, it is important to analyze the heavy metal concentrations in widely consumed fish species. The level of heavy metal contents in marine wild fish species from the Gabes gulf and particularly in Sfax coast are scarce.

The previous studies have been focused on the levels of heavy metal contamination in water and sediments (Gargouri et al. 2011; Serbaji et al. 2012). The present study is undertaken to evaluate the metal concentrations in the tissues (gills, liver, and muscle) of *L. aurata*, *D. annularis*, and *S. vulgaris* in water and sediments. The study is imperative in assessing polymetallic pollution since rapid urbanization and industrialization have adversely affected the south coast of Sfax.

Material and methods

Sampling

Water and sediment sampling

Water and sediment sampling were taken at 15 points in the south coast of Sfax and 10 points in Luza, located about 50 km north of Sfax. Luza was chosen as a control area, not directly subjected to sources of anthropogenic pollution (Fig. 1). Water samples were collected in 250-ml sterile bottles which were immediately stored at 4 °C for preservation until preparation and analysis. The samples were filtered through a 0.45-μm membrane and 25-ml aliquots from each one were prepared with 6 ml HNO_3 before analysis. Sediments were sampled with an Ekman bucket (10 cm diameter) at about 10/15 cm depth so as to obtain both sludge deposits. After collection, samples were wet sieved through a 5.0-mm pore-size polypropylene mesh with reagent grade water, for sediment fraction separation and elimination of detritus. The samples were then left to settle, and the water later decanted. Samples were dried in an oven at 80 °C for 3 days. The samples were later homogenized using a mortar and pestle and sieved to pass <63 μm (metals are most often associated with small grains; Morillo et al. 2004). The mortar, pestle, and sieve were cleaned before and after each sampling with 10 % redistilled HNO_3 and then rinsed with reagent grade water. One gram of each sediment sample underwent 3 h microwave digestion at 105 °C with 3 ml 6 M HNO_3 and 9 ml 5 M HCl.

Fish sampling

Three commercial fish species were purchased from fishing boats in two areas with different anthropogenic and natural impact in the south coast of Sfax and Luza, during December 2014. The collected species were the following: *Liza aurata*, *Solea vulgaris*, and *Diplodus annularis*. These fishes represent different biotopes and are economically important (Table 1). The set of investigated fishes was determined by the fact that they are the major edible fishes in Sfax. Collected fishes were wrapped in polyethylene plastic, put into an insulated cold container, and brought to the university laboratory where they were classified, weighed, measured by total length, and kept frozen at −20 °C until dissection.

For bioaccumulation analysis, the gills, liver, and muscle were taken from each fish (10 specimens of each species were analyzed). The fish in each composite sample had similar biometric parameters. The tissues were prepared for metal analysis as follows: muscle and liver were removed with stainless steel utensils, and gill filaments were removed from the gill arch scraped to remove overlying tissues. All were then dried at 80 °C for 3 days. One gram of each sample was

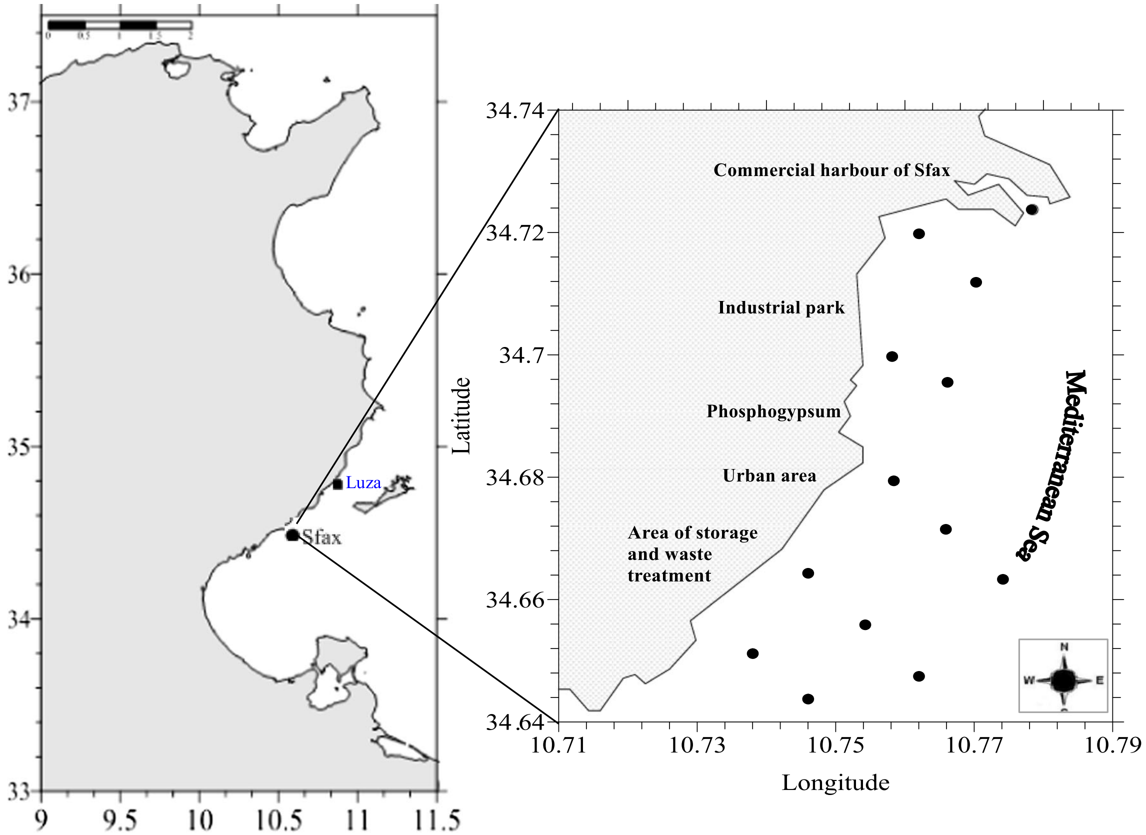

Fig. 1 Location map of the study area, south coast of Sfax. The *black circle* are the points of sediments samples

digested with 6 ml HNO_3 (65 %). Digestion was performed on a hot plate at 103 °C, for about 6 h.

Heavy metal analysis

The concentrations of heavy metals in water, sediments, and fish samples were determined with an atomic absorption spectrometer (AAS, Perkin Elimer). Detection limits obtained with AAS were 0.05 mg l^{-1} for Cd, Cu, and Pb and 0.5 mg l^{-1} for Ni. Metal concentrations were expressed either in milligrams per kilogram dry weight (D.W.) for sediment and fish or micrograms per liter in the water. To compare them with the FAO/WHO (1989, 2004) and WHO (1989) maximum permissible levels for human consumption, concentrations of metals in the liver and muscle were expressed in milligrams per kilogram wet weight (W.W.).

Transfer factor

The transfer factor (TF) in fish tissues from the aquatic ecosystem, which includes water and sediments, was calculated according to Kalfakakou and Akrida-Demertzi (2000) and Rashed (2001) as follows: where M_{tissue} is the metal concentration in fish tissue and $M_{sediment}$ is the metal concentration in sediment or in water.

$$TF = M_{tissue}/M_{sediment\ or\ water}$$

Metal load

The concentration of a single metal was divided by the mean concentration of that metal measured in tissues of fish from clean site (Luza zone). At each site, 10

Table 1 The ecological characteristics and recorded morphometric measures of examined fish species

Scientific name	English name	Family	Habitat	No. of samples	Length (cm)	Weight (g)
Diplodus annularis (Linnaeus, 1758)	Sparaillon	Sparidae	Benthopelagic	10	13.4 ± 0.46	25.80 ± 2.73
Solea vulgaris (Quensel, 1806)	Sole	Soleidae	Benthic	10	15.44 ± 0.47	34.25 ± 1.52
Liza aurata (Risso, 1810)	Mullet	Mugilidae	Pelagic	10	26.31 ± 0.49	60.74 ± 1.93

specimens of each species were captured, and metal levels were measured in the tissues. Standardized values were then added using the formula $ML_j = [\sum(C_{ij}/C_{ri})]/N$ where ML_j is the relative metal load in a tissue at site j, C_{ij} is the concentration of metal i at site j, C_{ri} is the concentration of metal i at the reference site, and N is the number of metals measured (Bervoets and Blust 2003). When $C_{ij} < C_{ri}$, the C_{ij}/C_{ri} was considered one. Thus the relative ML is a measure of the enrichment of the tissue with the measured metals, compared to those in fish from unpolluted sites. As a consequence, when no enrichment occurred, ML = 1.

Data analysis

Contour plots were made using the Surfer® 11 software. The results were expressed as means ± S.D. Data were statistically analyzed using Tukey's multiple range test to determine difference in means as indicated by different case letters in the descending order, a, b, and c at $P < 0.05$ using the Statistical Package for the Social Sciences software (SPSS, ver.20).

Result

Water analysis

The spatial distribution of heavy metal concentrations in the water is presented in Fig. 2a–f. Heavy metal concentrations in water samples decreased in the order Fe > Ni > Zn > Cu > Pb > Cd. The highest contents of Cd (0.21 μg l^{-1}), Cu (4.34 μg l^{-1}), Fe (30.74 μg l^{-1}), Ni (10.21 μg l^{-1}), Pb (3.43 μg l^{-1}), and Zn (5.78 μg l^{-1}) (Table 2) were found in station fronts to the points of the outfall of untreated domestic and industrial sewage and municipal leachate. The contents of Cu (stations 2, 5, 6, 8, 11, 12, 13, 14, and 15) (Fig. 2e) and Ni (stations 1, 2, 3, 5, 6, 11, 13, and 15) (Fig. 2f) exceeded the criterion continuous concentration (CCC) and the criteria maximum concentration (CMC), values of the USEPA water quality criteria (USEPA 1999).

Sediment analysis

Metal concentrations decreased in the following order: Fe > Zn > Pb > Ni > Cu > Cd. Averages, standard deviations,

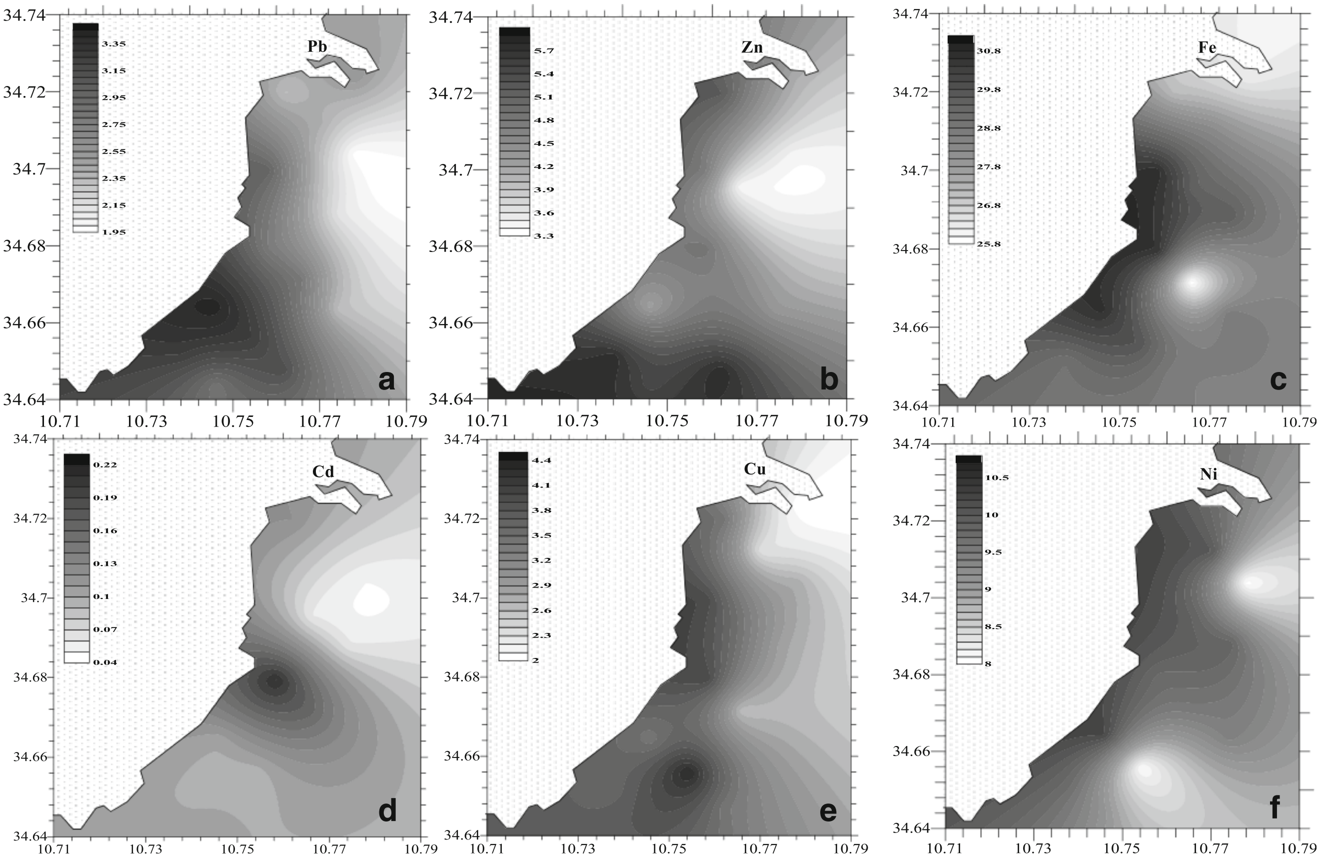

Fig. 2 Distribution maps of Pb (**a**), Zn (**b**), Fe (**c**), Cd (**d**), Cu (**e**), and Ni (**f**) concentrations (μg l^{-1}) in water

and ranges of heavy metal concentrations in superficial sediments, in the south coast of Sfax, are presented in Table 3. The metal concentrations (mg kg^{-1} D.W.) ranged from 65.06–151.50 for Pb, 47–546 for Zn, 50,564–11,956 for Fe, 4.42–7.92 for Cd, 8.23–28.56 for Cu, and 9.13–30.51 for Ni (Fig. 3a–f). The concentrations of the studied metals in sediments are higher than those in water. In an index of toxicity risk, Cd, Fe, Pb, and Zn suggest a potential toxicity on sediment-dwelling organisms in the south coast of Sfax. According to MacDonald et al. (2000), the reliability of the threshold effect concentration (TEC) and probable effect concentration (PEC) for assessing sediment quality conditions is determined based on their predictive ability. The average of Pb and Zn exceed the PEC, and the average of Cd and Fe exceed the TEC of quality assessment guidelines (SQGs) (Table 3).

Fish analysis

Concentrations of heavy metals (Cd, Cu, Fe, Ni, Pb, and Zn) in the muscle, liver, and gills of the *S. vulgaris*, *L. aurata*, and *D. annularis* are given in Table 4.

The highest Cu, Cd, Fe, Ni, and Zn concentrations were detected in the liver of *S. vulgaris*. The highest Pb concentration (mean ± SD; 5.06 ± 0.10 mg kg^{-1} D.W.) was recorded in gills of *S. vulgaris*.

The pattern of metal accumulation in three species was, in decreasing order:

- For *S. vulgaris*: Fe > Zn > Cu > Ni > Cd > Pb
- For *D. annularis*: Zn > Fe > Pb > Cd > Cu > Ni
- For *L. aurata*: Zn > Fe > Cu > Ni > Pb > Cd

The accumulation of metals in a single species showed significant inter-specific variations in all metals. However, it can be noticed that different tissues exhibited different patterns in metal accumulation.

Table 2 Heavy metals concentrations in the water (μg l^{-1}) in the south coast of Sfax (mean values and standard deviation are reported)

	Average	Standard deviation	Range	CMC[a]	CCC[a]
Cd	0.1	0.04	0.05–0.21	42	9.3
Cu	3.27	0.66	2.03–4.34	4.8	3.1
Fe	28.97	1.5	25.78–30.74		
Ni	9.44	0.73	8.03–10.21	74	8.2
Pb	2.72	0.44	1.98–3.43	210	8.1
Zn	4.59	0.78	3.43–5.78	90	81

[a] USEPA (1999)

Table 3 Heavy metals concentrations in sediments (mg kg^{-1} D.W.) in the south coast of Sfax (mean values and standard deviation are reported)

	Average	Standard deviation	Range	PEC[a]	TEC[a]
Cd	6.53	0.99	4.42–7.92	1	5
Cu	13.94	5.47	8.23–28.56	32	150
Fe	83,794	18,430	50,564–11,956	20,000	40,000
Ni	13.61	6.31	9.13–30.51	23	49
Pb	98.15	28.87	65.06–151.50	36	130
Zn	225.2	137.05	47–546.00	120	460

[a] MacDonald et al. (2000)

Transfer factor and metal load

TFs from water and sediments are given in Table 5. For Zn, TFs from the water were found to be greater than 1 in all tissue of all studied species. TFs from the water were found to be greater than those from sediments for all the analyzed elements. Except for Ni, all TF$_{water}$ exceeds one in gills and/or liver in the three studied fishes. TF$_{water}$ of Cd and Fe exceed one in the muscle *S. vulgaris* and *D. annularis*. These results confirm an intensive accumulation of those metals from water in these tissues. The highest TFs (organ/sediment ratio) were recorded in *S. vulgaris* tissues and the lowest were registered in *L. aurata* tissues.

Table 5 gives the mean relative metal load (ML) for the individual metals per tissue. Since the MLj the level of metal i in a tissue at site j, divided by the level of metal i at the reference site (Luza zone), MLj gives an indication of the enrichment of a certain metal in a tissue. The metal load exceeded one in the gills and/or liver of the studied species. The metal load analysis in the muscle remains important given that is destined for human consumption. The muscle ML*j* exceeds one in:

- *L. aurata* for Fe and Zn
- *D. annularis* for Cd, Fe, Ni, Pb, and Zn
- *S. vulgaris* for Cu, Fe, Ni, Pb, and Zn

Discussion

Heavy metals in water and sediment

The concentrations of the non-essential metals (Cd, Ni, and Pb) as well as essential metals (Fe, Cu, and Zn), that in high concentrations induce toxic effects in aquatic organisms (Oliveira et al. 2004), were quantified in the water and sediments of the south coast of Sfax. The metal pollution, in seawater, is due essentially to the presence of continuous discharge of local industrial and municipal effluent which

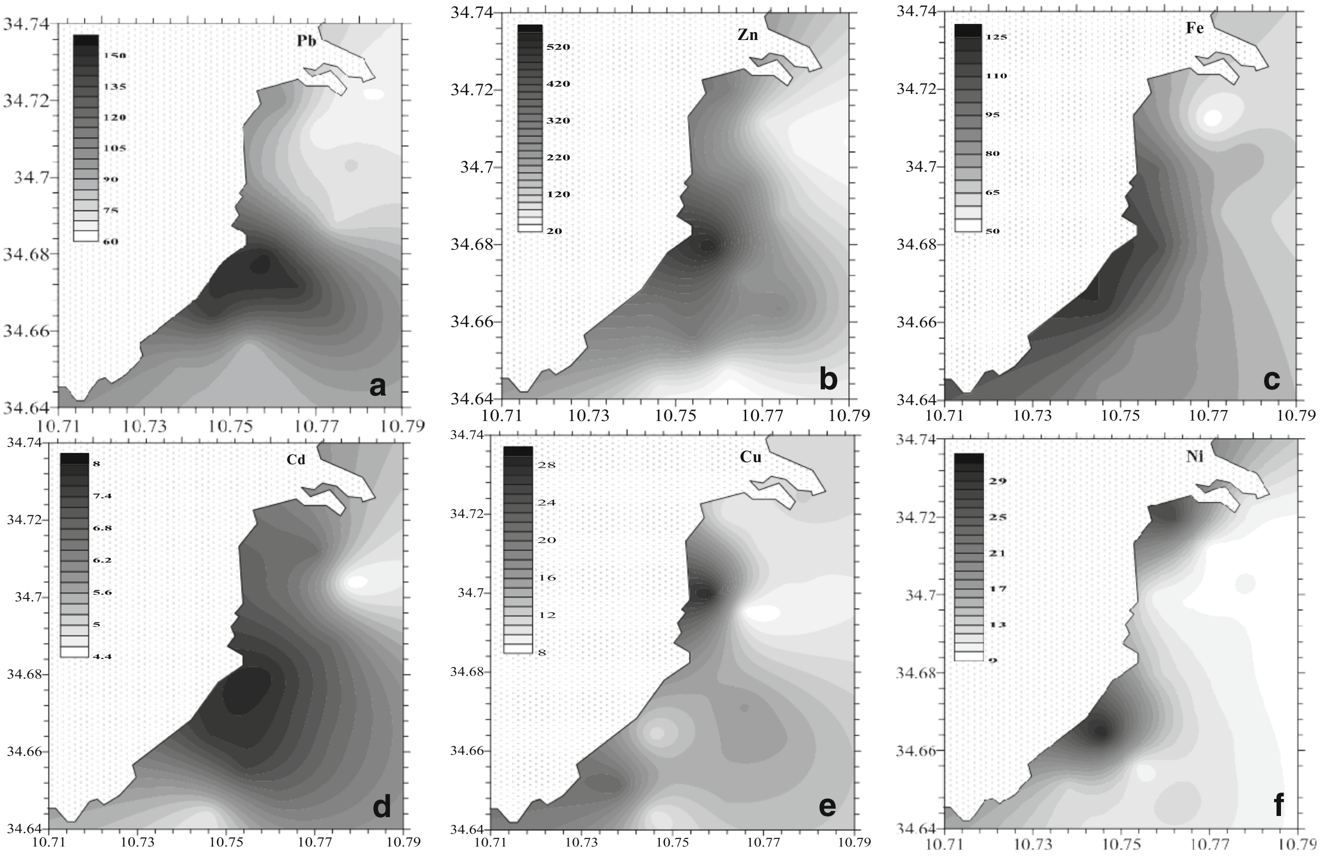

Fig. 3 Distribution maps of Pb (**a**), Zn (**b**), Fe (**c**), Cd (**d**), Cu (**e**), and Ni (**f**) concentrations (mg kg^{-1} D.W.) in surface sediments

contains a variety of organic and inorganic compounds, including heavy metals (Hamza-Chaffai et al. 2003; Smaoui-Damak et al. 2003; Banni et al. 2005, 2007; Messaoudi et al. 2009a, b; Gargouri et al. 2011). Indeed, the phosphogypsum stock is a product of the manufacture of phosphoric acid in the fertilizer industry which is not eliminated in the manufacturing process and thus, it enriches the seawater with dehydrated calcium sulfate and heavy metals as cadmium, zinc, chromium, copper, nickel, and lead (Degirmenci 2008; Kuryatnyk et al. 2008). The heavy metal concentrations in sediment were higher than those in water. The hypothesis that the sediment is the major sink and source of heavy metals in the marine

Table 4 Metal concentrations in tissues of *Diplodus annularis*, *Solea vulgaris*, and *Liza aurata* expressed in milligrams per kilogram D.W. (mean values ± standard deviations)

		Cd	Cu	Fe	Ni	Pb	Zn
Diplodus annularis	G	1.84 ± 0.05^{b}	0.34 ± 0.08^{b}	104.30 ± 0.08^{b}	0.35 ± 0.07^{a}	1.97 ± 0.05^{c}	137 ± 0.03^{b}
	L	3.01 ± 0.11^{c}	1.69 ± 0.02^{c}	240.80 ± 0.11^{c}	0.90 ± 0.14^{c}	0.28 ± 0.05^{b}	275 ± 0.10^{c}
	M	0.761 ± 0.02^{a}	0.11 ± 0.02^{a}	12.20 ± 1.09^{a}	0.66 ± 0.10^{b}	0.17 ± 0.01^{a}	135.77 ± 2.43^{a}
Solea vulgaris	G	3.69 ± 0.16^{b}	20.43 ± 0.16^{b}	343.22 ± 1.81^{b}	5.82 ± 0.08^{b}	5.06 ± 0.10^{c}	179.00 ± 0.94^{a}
	L	6.47 ± 0.26^{c}	40.81 ± 0.06^{c}	563.33 ± 0.03^{c}	6.42 ± 0.08^{c}	2.27 ± 0.72^{b}	284.90 ± 1.01^{c}
	M	1.03 ± 0.09^{a}	nd	43.50 ± 0.37^{a}	2.22 ± 0.49^{a}	1.85 ± 0.01^{a}	193.00 ± 0.45^{b}
Liza aurata	G	0.30 ± 0.03^{b}	0.04 ± 0.00^{b}	54.37 ± 0.06^{b}	0.08 ± 0.01^{a}	1.97 ± 0.35^{c}	171.2 ± 0.02^{c}
	L	0.55 ± 0.01^{c}	7.28 ± 1.12^{c}	197.92 ± 2.61^{c}	3.72 ± 0.84^{c}	0.57 ± 0.14^{b}	127.5 ± 0.06^{b}
	M	0.10 ± 0.05^{a}	nd	15.83 ± 0.06^{a}	1.23 ± 0.02^{b}	0.17 ± 0.05^{a}	106 ± 0.21^{a}

Different letters indicate significant differences between tissues a < b < c ($P < 0.05$)

Tissue concentrations found in dry weight were converted to wet weight by multiplying by a factor of 0.2 (considering average water content in fish tissues of 80 %)

nd not detected

Table 5 Transfer factor (TF) from sediment and water and metal load index

Species	Transfer factor and metal load	Tissues	Metals					
			Cd	Cu	Fe	Ni	Pb	Zn
Liza aurata	TF sediment	G	0.05	0	0	0.01	0.01	0.64
		L	0.08	0.54	0	0.31	0	0.48
		M	0.02	/	0	0.1	0	0.4
	TF water	G	2.88	0.01	1.83	0.01	0.62	32.3
		L	5.29	2.01	6.65	0.4	0.18	24.06
		M	0.96	/	0.41	0.13	0.04	20
	Metal laod	G	2.31	0.8	2.35	/	/	4.97
		L	5	1.78	3.5	/	/	4.84
		M	0.05	/	2.68	/	/	8.81
Diplodus annularis	TF sediment	G	0.28	0.03	0	0.03	0.04	0.51
		L	0.46	0.13	0	0.07	0	1.02
		M	0.12	/	0	0.05	0	0.51
	TF water	G	17.69	0.09	3.5	0.04	2.18	25.85
		L	28.94	0.47	8.09	0.1	0.09	51.89
		M	7.32	/	0.53	0.07	0.05	25.62
	Metal laod	G	1.45	4.68	3.01	1.94	3.38	5.16
		L	0.66	2.52	4.29	4.29	2.15	3.4
		M	1.9	/	7.57	6	3.4	3.27
Solea vulgaris	TF sediment	G	0.56	1.51	0	0.48	0.03	0.67
		L	0.99	3.02	0.01	0.53	0.01	1.06
		M	0.16	0.01	0	0.18	0.01	0.72
	TF water	G	35.48	5.63	11.53	0.63	1.58	33.77
		L	62.21	11.24	18.92	0.69	0.71	53.75
		M	9.9	0.03	1.46	0.24	0.58	36.42
	Metal laod	G	3.32	2.31	3.97	5.2	0.91	3.97
		L	2.15	2.24	1.95	2.41	17.46	3.89
		M	/	3.67	3.87	1.59	2.13	5.67

Average relative metal load (ML) is reported; mean concentration of a metal in a tissue divided by the concentration of that metal in that tissue of unpolluted site (Luza zone)

L liver, *G* gill, *M* muscle

environment (Connor 1984; Monikh et al. 2013; Naser 2013) was confirmed in the present study. Therefore, metals bound to the sediment might be released back into water columns with changed environmental conditions (Bryan and Langston 1992), resulting in potential long-term implication on human health and ecosystem.

Heavy metal concentrations in fish tissues

The study of heavy metal concentrations in fish tissues is crucial when it comes to human consumption. Considerable variations have been reported by several studies of heavy metal accumulation in various tissues of several fish species which may be related to metabolism and feeding patterns. The lowest concentrations of metals are shown in the muscle. The essential metals Cu, Fe, and Zn were accumulated mainly in the liver, while Pb exhibited the highest concentrations in the gills.

Differences in metal accumulation in fish tissues are conditioned by some factors depending on the intensity of fish metabolism. After incorporation into the fish body, heavy metals were distributed among different tissues via a process that depends on biological needs (Zubcov et al. 2012).

Cadmium is a non-essential highly toxic and ecotoxic metal (Stancheva et al. 2013). It may accumulate in the human body and may induce skeletal, hepatic, renal, pulmonary, and reproductive effects and cancer (FAO/WHO 2004). The highest amount of cadmium was found in the liver of *S. vulgaris* (6.47 ± 0.26 mg kg^{-1} D.W.) and the lowest amount of Cd was found in the muscle of *L. aurata* (0.10 ± 0.05 mg kg^{-1} D.W.). Our experimental study revealed that, except for liver of *S. vulgaris* (1.30 mg kg^{-1} W.W.), Cd concentrations in the selected fishes from the south coast of Sfax were below the permissible limits 1 mg kg^{-1} W.W. set by world human organizations (FAO/WHO 1989), but a long period of accumulation of Cd in fish may pose health hazards. Copper is an essential part of several enzymes and is necessary for the synthesis of hemoglobin (Sivaperumal Sankar et al. 2007; Monteiro et al. 2009). However, high intake of Cu has been recognized to cause adverse health problem (Flemming and Trevors 1989; Gorell et al. 1997; Hernández et al. 2006). Cu concentrations were below the detection limits in the muscle of *L. aurata* and *D. annularis*. The highest copper levels in fish species were found as 40.81 ± 0.06 mg kg^{-1} D.W. in the liver of *S. vulgaris*. Cu concentrations in all tissues of studied species were below the maximum permitted copper level, 30 mg kg^{-1} W.W. established by FAO/WHO (1989) and WHO (1989). Iron is an essential element and has a vital role in the enzymatic and respiratory processes of fish (Paez-Osuna and Ruiz-Fernandez 1995); however, it is toxic at high concentrations (Cairo et al. 2006). In the present study, Fe concentrations ranged from 563.33 mg kg^{-1} D.W. (liver, *S. vulgaris*) to 12.20 mg kg^{-1} D.W. (muscle, *D. annularis*). Fe concentrations in the liver (112.66 mg kg^{-1} W.W.) of *S. vulgaris* exceed the WHO permissible level 100 mg kg^{-1} W.W. for safe human consumption (WHO 1989). Lead is a non-essential element, and it is well documented that Pb can cause neurotoxicity, nephrotoxicity, and many other adverse health effects (García-Lestón et al. 2010). The higher average concentration is measured in the gills (5.06 mg kg^{-1} D.W.) for *S. vulgaris*. This concentration (1.01 mg kg^{-1} W.W.) is two times higher than the maximum permitted level 0.5 mg kg^{-1} W.W. (FAO/WHO 1989). Nickel is an essential element, but it is a potentially toxic element and can cause variety of pulmonary adverse health effects, such as lung inflammation, emphysema, tumors, and fibrosis (Forti et al. 2011; Sfakianakis et al. 2015). In the present investigation, the highest amount of nickel was found in the gills and liver of *S. vulgaris*, and it is below the detection limit in the muscle of *L. aurata*. The Ni concentrations in the gills and liver of *S. vulgaris* (1.16 and 1.28 mg kg^{-1} W.W., respectively) exceed the maximum permitted level 0.5–1 mg kg^{-1} W.W. (FAO/WHO 1989). Zinc is an essential element, known to be involved in most metabolic pathways in humans, such as the immune system, neurotransmission, and cell signaling (Tuzen 2009), but at high levels, they can be toxic (Niyogi and Wood 2006). The concentrations of Zn ranged from 284.90 ± 1.01 mg kg^{-1} D.W. in the liver of *S. vulgaris* to 106 ± 0.21 mg kg^{-1} D.W. in the muscle of *L. aurata*. Except for Zn concentration in the liver of *S. vulgaris* (56.98 mg kg^{-1} W.W.), the amount of Zn did not exceed the FAO/WHO permissible level 40 mg kg^{-1} W.W. for safe human consumption (FAO/WHO 1989). As a variety of metal load (Ni, Pb, and Zn) was found in *S. vulgaris*, a high metal load was likewise found in *D. annualris* and *L. aurata*. It thus appears that for good bio-monitoring of environmental pollution, different species must be analyzed.

Variations in organs ability to accumulate metals

Distribution of pollutants among the various organs within an organism is heterogeneous, but rather they accumulate in specific target organs (Terra et al. 2007). The liver has been recommended by many authors as the best environmental indicator, since the amount of pollutants in fish liver is directly proportional to the degree of pollution in the aquatic environment (Agah et al. 2009; Messaoudi et al. 2009a; Yilmaz 2009; Tapia et al. 2012). The liver is targeted due to its role in trace metal storage, redistribution, detoxification or transformation, and importance to individual fish health (Cizdziel et al. 2003; Licata et al. 2005; Hasyimah et al. 2011). Fish respond to metal exposure by producing metallothioneins (MTs), particularly in the liver (Kargin 1998). The accumulation of essential metals in the liver is likely linked to its role in metabolism (Zhao et al. 2012); high levels of Zn and Cu in hepatic tissues are usually related to a natural binding protein such as MTs (Qadir and Malik 2011) which act as an essential metal store (Zn and Cu) to fulfill enzymatic and other metabolic demands (Roesijudi 1996; Amiard et al. 2006). In the same way, Fe tends to accumulate in hepatic tissues due to the physiological role of the liver in blood cells and hemoglobin synthesis (Korkmaz Görür et al. 2012; Omar et al. 2014). On the other hand, the liver also showed high levels of non-essential metals such as Cd and Ni; liver tissues are expressed to be the main site of trace metal detoxification within fish. High concentrations of Cd could be explained by the ability of this element to displace the normally MT-associated essential metals in hepatic tissues (Amiard et al. 2006). Similar results of high Fe, Zn, Cu, and Cd in the liver were observed in many field studies (Dural et al. 2007; Zhao et al. 2012; El-Moselhy et al. 2014). Gills are the main route of metal ion exchange from surrounding seawater (Romeo et al. 1999; Farkas et al. 2003; Qadir and Malik 2011) as they have very large surface areas that facilitate rapid diffusion of metals (Farkas et al. 2003; Dhaneesh et al. 2012). High concentration of various metals in the gills could be due to the element complexing with the mucus, which has an affinity to be bound with metal ions (Usero et al. 2004; Doraghi et al. 2011). The lowest metal values were found in the muscle; this may be due to the fact that the muscle is not an active tissue in the accumulation of heavy metals (Karadede et al. 2004; Tekin-Özan and Kir 2008; Ebrahimpour et al. 2011). Although fish muscle tended to accumulate low concentrations of metals, it is important to compare these to known safety levels because the muscle constitutes the greatest edible part of the fish (Yilmaz 2003; Storelli et al. 2006; Castro-González and Méndez-Armenta 2008; Zhuang et al. 2013; Ben Salem et al. 2014). In the present study, the amounts of studied heavy metals are higher than those reported by other studies in the Mediterranean Sea. Thus, the concentrations of Cd, Pb, Fe, Cu, Ni, and Zn in the muscle of *L. aurata* are higher than those found by Ketata Khitouni et al. (2014, Gabes gulf area), Ersoya and Celik (2009, Iskenderun Bay). *D. annularis* show high concentrations of Pb and Fe than those reported by Ersoya and Celik (2009, Iskenderun Bay). For *S. vulgaris*, Cd concentrations in the gills and liver were slightly higher than those founded by Barhoumi et al. (2009, Gabes gulf area).

Inter-specific variations in metal accumulation

Previous researchers highlighted that there were differences between metal levels in studied species and concluded that this aspect was very significant from both an ecotoxicological view point and concerning public health risks associated with their consumption (Storelli et al. 2006). It is very difficult to compare metal concentrations, even between the same tissues of two different species, because of different feeding habits (as *S. vulgaris* feed on a wide variety of prey types, copepods, polychaetes, algae, mollusks, seagrasses, and amphipods with frequent quantities of sediments (El-mor and Ahamed 2008)), *L. aurata* feed mostly on zooplankton and *D. annularis* feeding preferentially on gastropods and small crustaceans (Pita et al. 2002), ecological needs, metabolisms, types of tissues analyzed, differences in the aquatic environment concerning the type and levels of water pollution, and other factors (Valle et al. 2003; Oliveira et al. 2010; March et al. 2011; Kucuksezgin et al. 2014).

In our findings, the concentrations of trace metals in fish samples indicate that *S. vulgaris* seemed to be more contaminated than were other fish species, followed by *D. annularis* and *L. aurata*. Unlike others, *S. vulgaris* appeared to tend to concentrate large amounts of Cu, Ni, and Pb, demonstrating a potential as a bio-indicator of pollution of the coastal ecosystems. These observations are mainly due to different fish habitats (*S. vulgaris* is a benthic species while *D. annularis* and *L. aurata* are benthopelagic and pelagic species, respectively) and surrounding ecosystem status. In agreement with Bustamante et al. (2003) who has suggested that benthic fish are likely to have higher heavy metal concentrations than fish inhabiting the upper water column because they are in direct contact with the sediments. One important implication of our findings is related to the levels of the essential elements, normally considered as essential to living organisms, although almost all become toxic when present at high levels. Indeed, several attentions may be given to the non-essential elements to prevent toxic effect and ensure safe human consumption.

Transfer factors and metal load

A TF greater than 1 indicates bioaccumulation (Kalfakakou and Akrida-Demertzi 2000; Rashed 2001). Zinc was found to be highly accumulated in the fish species of the present study, and it is attributed to the high amounts of domestic and industrial wastes from the surrounding inputs. The high TF_{water} of Zn (53.75) and Cd (62.21) signify slow accumulation and their potentiality for chronic effects and accretion in the food chain of organisms (Jayaprakash et al. 2015; DeForest et al. 2007).

According to the TF_{water} values, the species could be ordered as *S. vulgaris* < *D. annularis* < *L. aurata*. According to Dallinger (1993), the fish species can be classified based on the $TF_{sediment}$ values which include the macroconcentrator ($TF_{sediment} > 2$), microconcentrator ($1 < TF_{sediment} < 2$) and deconcentrator ($TF_{sediment} < 1$). Our results revealed that *D. annularis* and *L. aurata* are deconcentrators and *S. vulgaris* can be considered as macroconcentrator. Thus, the species *S. vulgaris* with the highest (3.02) $TF_{sediment}$ would be considered as a potential bio-indicator in the south coast of Sfax for the assessment of environmental pollution status. The metal load (ML) exceeding one, namely in the muscle of the studied fish. The highest ML values were recorded in gills of *D. annularis* for Zn (5.16), in the liver of *S. vulgaris* for Pb (17.46), and in the muscle of *L. aurata* for Zn (8.81). These results highlight the enrichment of the fishes from the south coast of Sfax on heavy metals especially on Pb and Zn in comparison with Luza zone.

Conclusion

The south coast of Sfax as an important ecosystem has received an increased attention due to its obvious role on the fishery. This study evaluated the levels of metal accumulation in water and sediment and gills, liver, and muscle of the three fishes *S. vulgaris*, *L. aurata*, and *D. annularis*. Concentrations in sediment were generally higher than in water, and based on the comparison with sediment quality guidelines (SQGs), Cd, Fe, Pb, and Zn exceed PEC values causing adverse effects on sediment-dwelling organisms. Bioaccumulation of heavy metals in seafood is a major health concern worldwide. The metal content in fish liver and gills was considerably higher than in muscle. Concentrations of Fe and Zn were higher than Cd, Cu, Pb and Ni in tissue analyses. Given that concentrations in fish tissues reflect the environmental conditions in fish habitat (except for Cd, Pb, and Ni), we can presume that the south coast of Sfax is heavily loaded with Fe and Zn. Indeed, the metal load index indicates that the study site is submitted on Pb and Zn pollution. Thereby, the fish resources were threatened by metallic pollution. Our results showed that metal accumulation varied between species depending on species-specific factors like feeding behavior, fish habitat, and surrounding ecosystem status that caused variation in metal accumulations between fish. Further investigations remain necessary to identify the probable health hazards for the people in the Sfax city who consume more seafood than an average inhabitant.

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