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Assessment of metallic contamination in sediment and mullet fish (*Mugil cephalus* Linnaeus, 1758) tissues from the East Algerian coast

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Abstract

The present work was designed to assess the contamination level in the coastal of Annaba, following the spectrophotometric determination of the level of some metallic elements (Fe, Cu, Pb, Zn, Ni and Cd) in the sediments along an increasing bathymetric gradient (10 m and 20 m), as well as in the biological indicator grey mullet (*Mugil cephalus*) muscle. During the winter period (2014), 12 surface sediment samples, and a total of 24 fish were collected. Once the samples are dried, crushed and sieved, 0.5 g dry weight of each sample was added to concentrated acids. The results showed that the levels of some metals are superior to the recommended guideline values, and consequently the sediment of this bay is contaminated by iron, lead and copper. The contamination index (*CI*) showed a general tendency in the concentration of the studied metals as follows: Fe > Pb > Cu > Zn > Ni > Cd, since the most studied metals occurred at higher concentration in depth (20 m).

However, the average concentrations of metals in fishes were found to be as the following order: Fe > Zn > Pb > Cu > Ni > Cd. The consumption of fish from some contaminated sites can be dangerous because concentrations of lead, cadmium and zinc exceed the international standards. Moreover, the strong positive correlation observed between the metallic elements of sediments and fishes confirm that these metals resulted from the similar sources of the anthropic activities, such as domestic, port, industrial and agricultural waste discharges. This is confirmed by the determined of pollution load index (*PLI*) parameter. Conclusively, a regular monitoring program of heavy metal is recommended for protecting these organisms, and to reduce the environmental risks.

Key words: *fish, heavy metals, sediment, toxicity, water depth*

INTRODUCTION

The metallic contamination of the aquatic environment has become a world concern, due to their toxicity, bioaccumulation and the harmful effects on aquatic living organisms (fauna, flora and microorganisms), in which the metallic residues accumulate in their soft tissues. Human are exposed to these toxic metals through consumption of contaminated water and food also can be targeted the metallic residues affect the aquatic life [YAKINOV *et al.* 2007]. The pollution by trace metals, that considered to be as the normal components of the environment at trace levels [BRYAN 1984], but they may become toxic above a certain threshold concentration [KUCUKSEZGIN *et al.* 2006].

Heavy metals are pollutants whose harmfulness is related to their persistence and toxicity, some of them are known as essential elements (zinc, iron and copper) that play a crucial role in the biological processes, and others considered as non-essential elements (no role in the biological processes) able to induce toxic effects when their concentration exceeds certain threshold of acceptability [CHEN *et al.* 2016; LAFABRIE *et al.* 2007]. Because they neither be metabolized not be excreted by the biological processes, and therefore they accumulate in various components of the ecosystem (water, sediment, flora, and fauna). Due to the cumulative effects of trace metallic elements in plants and trophic chain, serious environmental and even human health problems are strongly associated to metal contamination [LENOBLE *et al.* 2013]. Moreover, toxic metals cause harmful effects on physiological functions, individual growth rates, reproduction and mortality of aquatic living organisms [RUQIA *et al.* 2015; YUJUN *et al.* 2011]. The uptake of heavy metals in fish follows three possible pathways: the body surface, the gill or the digestive tract, following three exposure sources: food, water and sediment. Importantly, the sediment is a milieu in which the micropollutants may be accumulated and fixed for a long-term period, and thus their analysis is valuable tool for the determination of pollution level [BUGGY, TOBIN 2008]. Noteworthy, the coastal areas known as urbanized and industrialized areas are strongly exposed to high waste discharges of heavy metals due to anthropic activities [PEKEY 2006]. In these regards, the study was focused on the Annaba Gulf (North-East of Algeria) known as one of the principal gulfs in Algeria. The gulf is exclusively fed from the water of Seybouse River, which is considered the vital source for the north-eastern regions of Algeria, however, it undergoes serious environmental pollution problems due to permanent urban and industrial discharges [BELABED *et al.* 2017]. The aim of our study, therefore, was to evaluate the chemical pollution level in Annaba Gulf, in order to understand its health state, and subsequent to find out the best management ways and protection of its coastal zones. Here, we carried out the estimation of the contamination by the six heavy metals, respectively, Zn, Cu, Pb, Fe, Ni and Cd in the sediments using the contamination index and the biological indicator mullet (*Mugil cephalus*), known as bio accumulative species for metals, as well as widely used species as a best indicator for metal pollution.

MATERIALS AND METHODS

DESCRIPTION OF THE STUDY ZONE

The study zone is located in the extreme East part of the Algerian coast, 600 km from Algiers and 100 km from the Tunisian border. The Annaba Gulf of is a wide-mouthed bay, open to the Mediterranean Sea on the North, extending from Cap-Rose (8°1' W and 36°58' N) in the East to Cap de Garde (7°47' E and 36°58' N) in the West, with a distance of 40 km between them. The Seybouse River in the South-East, the second longest river in Algeria (with a catchment basin of about 6470 km^2) located in the South-East part, and the Mafrag River in the East drain into the gulf [BELABED *et al.* 2013]. The two rivers receive agricultural water discharges from cereal farming,

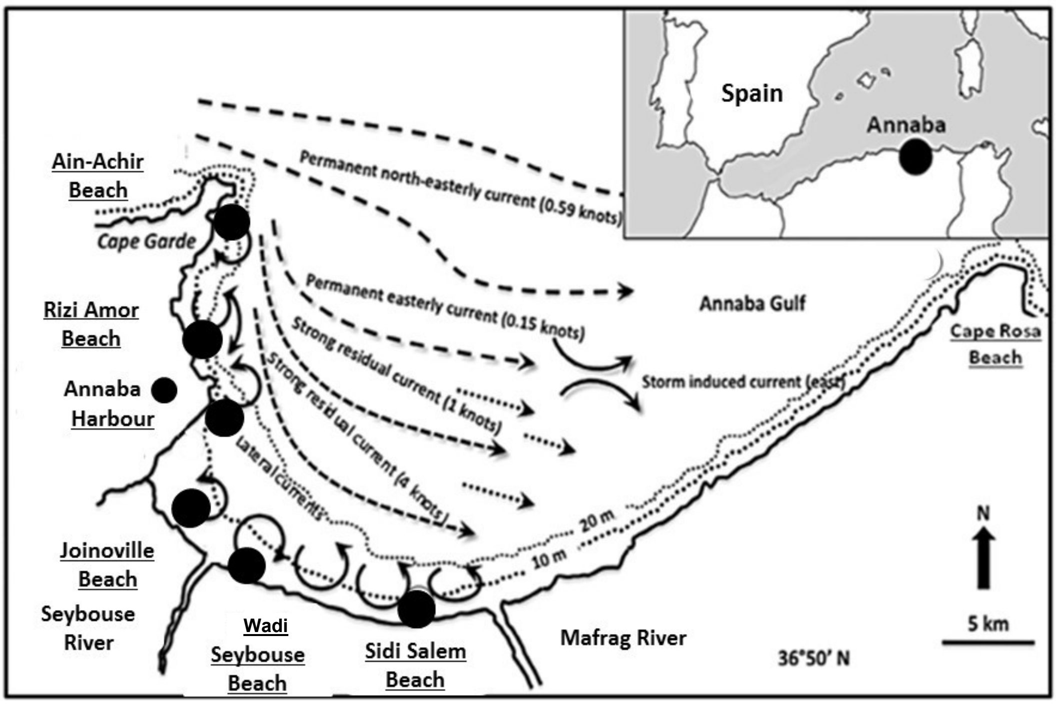

Fig. 1. Location of the sampling sites along of Annaba Gulf; source: BELABED *et al.* [2013], modified

market gardening and arboriculture, and domestic releases from important conurbations [KHELIFI-TOUHAMI *et al.* 2006] and untreated sewage [ABDENNOUR *et al.* 2000] contributing to sediment contamination by heavy metals. Moreover, discharges from industries, which settle in the lower part of the Meboudja River, the final tributary of the Seybouse River, are important source points of heavy metals measured in the Annaba Gulf superficial sediments. The gulf has a Mediterranean climate characterized by an annual mean temperature of 18°C and precipitations ranging from 650 to 1000 mm, mainly distributed in winter (60–70% of total precipitation) and, therefore, winters are cold and humid, since summers are worm and dry. Additionally, the gulf is subjected to a dominant sea-breeze with a speed reaching 6 $m \cdot s^{-1}$ from the North–North-East (30%), and, to a lesser extent, from the north (13%) and the west (10%) [DEBIECHE 2002]. The maximal depth of the gulf is 65 m.

The sampling points were chosen in relation to locations of human activities that may be the source of pollution, namely the proximity of constructions, pipe spillway of wastewater, as well as the industrial zone (Fig. 1, Tab. 1).

Table 1. Main industries discharging wastes into Annaba Gulf [ABHA 1999].

Station	Coordination	Source of pollution	Type of pollution
Sidi Salem	36°51'42" N 07°46'59" E	urban wastes	domestic waste water
Wadi Seybouse	36°52'03" N 07°46'29" E	urban and industrial wastes: (ORELAIT, E.N.C.C, Ferrovial, Tils-Granito)	domestic waste water waters of cooling industrial waste waters
Joinoville	36°52'14" N 07°46'10" E	urban and industrial wastes (Asmidal)	domestic waste waters industrial waste waters waters of cooling
Harbour	36°53'40" N 07°46'30" E	urban and industrial wastes: (Véadu, E.N.C.G, O.N. food of livestock n°1)	domestic waste waters industrial waste waters
Rizi Amor	36°55'39" N 07°45'16" E	urban wastes	domestic waste waters
Ain-Achir	36°57'59" N 07°47'42" E	there are no wastes	–

Source: own elaboration.

COLLECTION AND PREPARATION OF SAMPLES

The Figure 1 shows the location of six sampling stations: Sidi Salem Beach (St1) (36°51'42" N and 07°46'59" E), located in the eastern part of the study area, close to the outlet of the Mafrag River; Wadi Seybouse Beach (St2) (36°52'03" N and 07°46'29" E), known as Wadi Seybouse in referring to Seybouse River that flows into it, receives agricultural, industrial and urban waste from the west valley of the city of Annaba; Joinoville Beach (the industrial zone of Asmidal) (St3) (36°52'14" N and 07°46'10" E), located in the North-East of the city of Annaba in the municipality of El Bouni, near Annaba harbour; harbour station (St4) (36°53'40" N and 07°46'30" E), is located in the centre of the city of Annaba, ensures the transit of passengers and merchandise, it is subject to various sources of pollution; Rizi Amor Beach (St5) (36°55'39" N and 07°45'16" E), is part of the west coast of Annaba Gulf, receives also urban waste; Ain-Achir Beach (St6), is part of the extreme west of the coastal zone, this point is located between 36°57'59" N and 07°47'42" E and supposed to be exposed to no sources of pollution (Tab. 1).

The twelve (12) samples of surface sediments were collected from a river depth of 10 m and 20 m, within winter period (25.12.2014). Samples of 250 g of surface sediments were scraped from a polyethylene shovel. The samples were taken in polyethylene containers, transported in icebox to laboratory, and afterwards they were stored at 4°C until being analysed [PNUE 1985]. The *Mugil cephalus* fish sampling is also carried out during a winter season at the same period for the same stations situated in the west section of the Gulf. A total of 24 fishes, grouped as 4 fishes per site, along with average total length (*TL*) of 25.7 ± 2.69 cm (22.7 ≤ *TL* ≤ 31.3 cm) and average total weight (*TW*) of 87.05 ± 5.62 g (77.45 ≤ *TW* ≤ 95.55 g) were studied.

ANALYTICAL PROCEDURE AND ANALYSIS

All the laboratory plastics and glassware were cleaned by soaking overnight in nitric acid solution of 10%, followed by rinsing with bidistilled water. The dried sediments placed in stove at 80°C during 48 hours were crushed in agate mortar, then sieved into fine powder, and only the particles smaller than 65 μm in diameter were collected for later use. Indeed, the fine particles are generally rich in pollutants, and contain a large part of clay levels. A quantity of 0.5 g of dry weight sediments was mixed with 0,01 dm^3 of HNO_3 (ultra-pure) and 0.05 dm^3 of $HClO_4$ (ultra-pure) at a temperature of 150°C during 16 hours, in order to obtain a good digestion. The mineralized residues were subjected to a consecutive rinsing in bidistilled water, filtered with Whatman filter (0.45 mm), and afterwards the solution was completed to a final volume of 0.1 dm^3 with distilled water [SALMONS, FÖRSTNER 1984].

The concentration (C, in $mg \cdot kg^{-1}$) of metallic element in the sediment is obtained according JOANNY *et al.* [1983] as follow:

$$C = CE \cdot V/M \tag{1}$$

Where: V = the final volume of solution after digestion (cm^3); M = mass of the mineralized sediment (kg); CE = the concentration read from the calibration curve ($mg \cdot (0.001\ dm^3)^{-1}$).

Each sample of fish (muscle) was carefully dissected. To prevent the metal contamination, special care has been taken into account, and the tissues were dissected with special ceramic knife, scissors and plastic clips. After that fish muscle samples from each individual were cleaned with bidistilled water, cut out into small pieces (2–3 cm) and dried overnight in oven at 65°C [UNEP 1982]. Then were pulverized and sieved through 1 mm of size mesh, the dried fish muscles were digested as described elsewhere [RAHMAN *et al.* 2012]. Approximately 0.5 g of tissues as a dried powder was added to concentrated nitric acid (0.004 dm^3) and concentrated sulfuric acid (0.0025 dm^3) [UNEP 1982; 1984]. Then the mixture was heated slowly during 20 min at 130°C. As a result, the mixture was filtered by Whatman filter (0.45 mm), and thereafter the solution was completed to a final volume of 0.01 dm^3 with distilled water. The detection of heavy metals (Zn, Cu, Pb, Fe, Cd and Ni) in the all samples (sediments and fish) was carried out according flame atomic absorption spectrometric method as described by AMINOT and CHAUSSEPIED [1983].

All elements were afterwards expressed as $mg \cdot kg^{-1}$ of dry weight. The detection limits of heavy metals in sediment and fish samples for cadmium (Cd), copper (Cu), iron (Fe), nickel (Ni), lead (Pb) and zinc (Zn) were 0.02, 0.03, 0.04, 0.06, 0.06 and 0.05 $mg \cdot kg^{-1}$, respectively. The precision of the method was checked by several measurements on the standard reference materials of the International Agency for Atomic Energy (IAAE): IAEA-407 (fish tissue) and IAEA-SL-1 (riparian sediment). Recoveries were above 90% for all the heavy metals measured. Results are indicated in Table 2.

Table 2. Certified and observed values of trace metal concentrations in reference materials (biota – IAEA-407, sediment – IAEA-SL-1), in $mg \cdot kg^{-1}$ dry weight ± standard deviation

Metal	Biota		Sediments	
	certified acc. to IAEA-407	observed	certified acc. to IAEA-SL-1	observed
Zn	67.1 ± 3.8	65.5 ± 0.7	223 ± 10	228.04 ± 14.08
Cu	3.28 ± 0.40	3.52 ± 0.18	30.0 ± 5.6	34.03 ± 6.39
Pb	0.12 ± 0.06	0.137 ± 0.006	37.7 ± 7.4	37.29 ± 8.95
Cd	0.189 ± 0.019	0.187 ± 0.003	0.260 ± 0.050	0.26 ± 0.06
Fe	n.r.	n.r.	6.74 ± 0.17	6.54 ± 0.27
Ni	n.r.	n.r.	44.9 ± 8.0	49.13 ± 7.47

Explanation: n.r. = non-certified values for the concentrations of some elements in IAEA-407.
Source: own elaboration.

QUALITY STANDARDS

From a regulatory viewpoint, the metallic pollution treatment in the surface sediments and the fish tissues differs from one country to another. In Table 3, we noticed the allowable limits linked to the superficial marine sediments known by the French laws [ABRMC 1991], as well as at the fish tissue levels compared to allowable maximal limits recommended by the world health organization [WHO 2004] and the Environmental Protection Agency of USA [USEPA 2002].

Table 3. International security standards of heavy metals in sediments and fish

Heavy metal	Sediments acc. to ABRMC [1991]	Fish muscles acc. to	
		WHO [2004]/ USEPA [2002]	CE [2006]
	$mg \cdot kg^{-1}$ dwt		
Ar	10	–	–
Cd	0.6	1	0.05
Cu	26	30	–
Cr	45	1	–
Pb	22	2	0.30
Zn	88	100	–
Fe	2000	–	–
Mn	400	1	–
Ni	45	0.5–1	–

Source: own elaboration acc. to literature.

In the case of French standards, the evaluation of the contamination index values (*CI*, or guideline) used in the present study for the surface sediments are comparable to those of the Agency of Rhone-Mediterranean and Corsica Basin [ABRMC 1991]. Interestingly, the contamination index (*CI*) represents the ratio between the reference value of given heavy metal and its level in the studied sediment.

If *CI* is inferior to 3, the sediment is considered as class 1 (normal zone), the sediment is of class 2 (polluted zone) when *CI* is between 3 and 10, since the sediment belongs to class 3 (risk zone) when *CI* is superior than 10.

The evaluation of contamination degree was also done by using the pollution load index (*PLI*). The later has been widely used to evaluate the contamination level and the pollution in the estuarine and coastal sediments.

The following equation to calculate *PLI* has been developed by TOMLINSON *et al.* [1980]:

$$PLI = (FC_1 \cdot FC_2 \cdot FC_3 \cdot \ldots \cdot FC_n)^{1/n} \tag{2}$$

$$FC = C_{metal}/C_{background} \tag{3}$$

Where: FC = the contamination factor, n = the number of the analysed elements, C_{metal} = the concentration of element in the sediment, $C_{background}$ = the geochemical background value of the element.

The mean comparisons of the spatial data of heavy metal concentrations in the superficial sediments and fish muscle were tested by ANOVA. The relation between the analysed elements (Zn, Cu, Pb, Fe, Ni and Cd) in the sediment and *M. cephalus* of Annaba Gulf was tested by using the Pearson correlation coefficient, with a significance levels: $p \le 0.05$, $p \le 0.01$ and $p \le 0.001$.

RESULTS AND DISCUSSION

METALLIC LEVELS IN THE SEDIMENT

The results relative to the trace element concentrations in superficial sediments of the study zone are given in Figure 2. The calculated contamination index

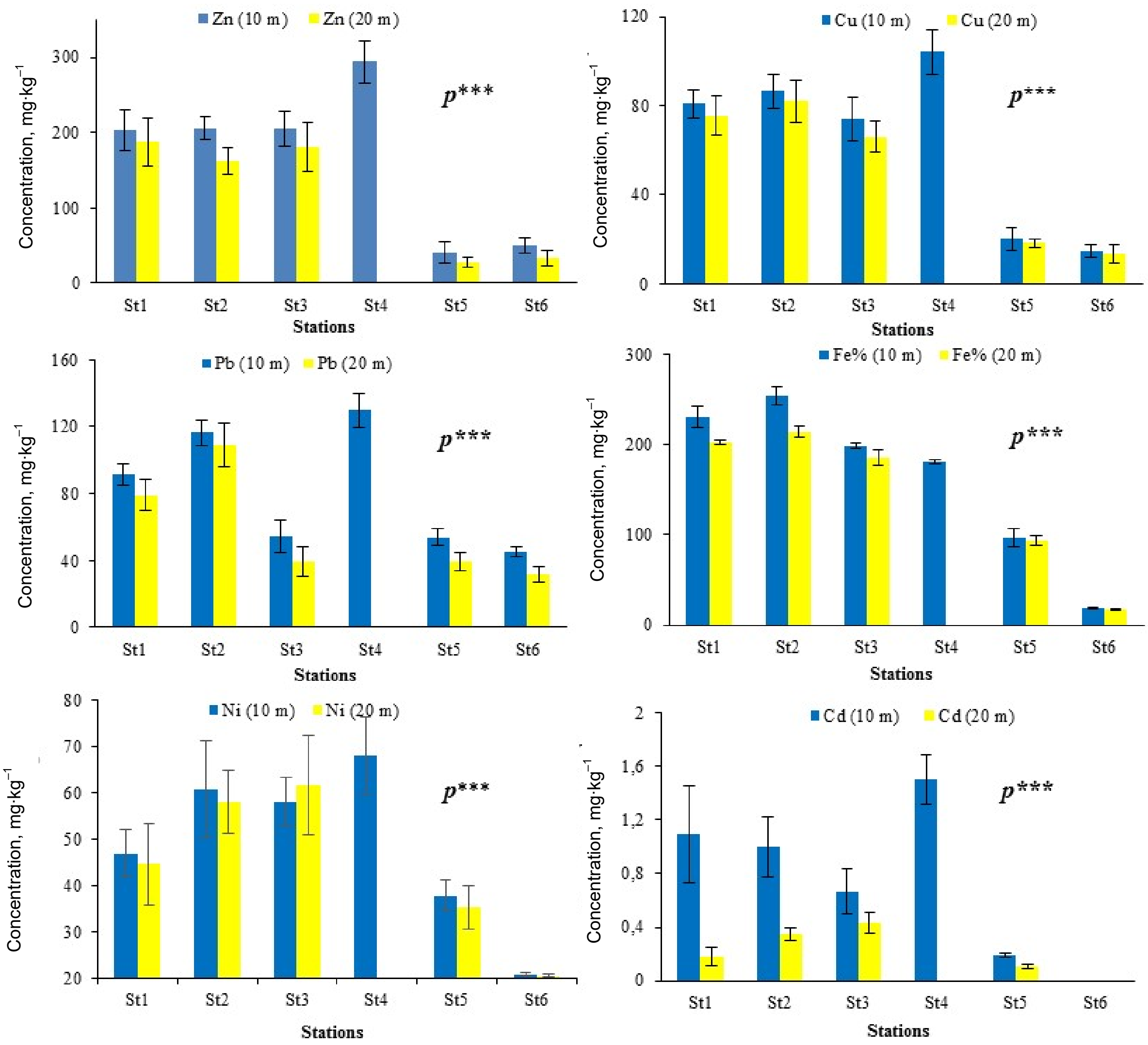

Fig. 2. Average concentrations of metals in sediments (mg·kg^{-1} dry weight) of the six selected stations: Sidi Salem Beach (St1); Wadi Seybouse Beach (St2); Joinoville Beach (the industrial zone of Asmidal) (St3; harbour station (St4); Rizi Amor Beach (St5); Ain-Achir Beach (St6); $p \leq 0.001$; source: own study

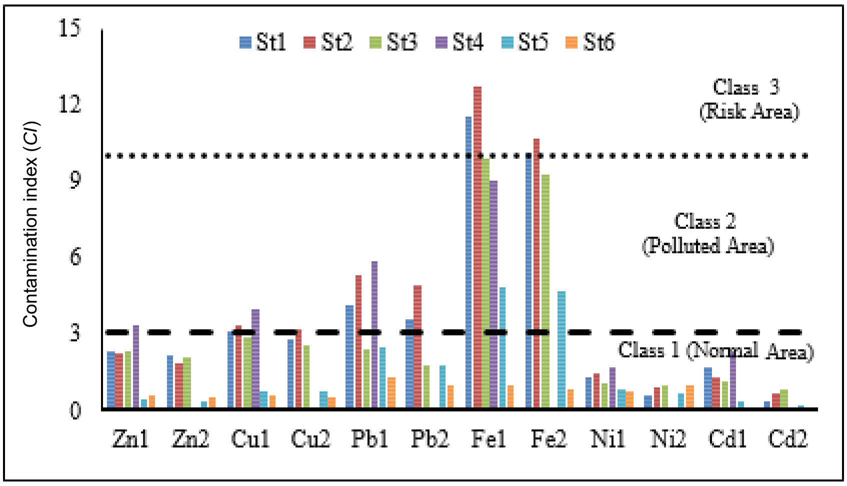

Fig. 3. Variations of the contamination index (*CI*) of various metals in the Annaba Gulf (1 = 10 m; 2 = 20 m); St1–St6 as in Fig. 2; own study

(*CI*) for every analysed metal in the sediments of six stations varies generally from one station to another (Fig. 3). Since the iron is an essential element that may be toxic at higher concentrations, iron is found to be an abundant element in the studied stations with mean values of 22 680 ± 653 mg·kg^{-1} with a *CI* of 11.34, confirming that iron is an element belongs to class 3. The sediment of station 1 (21 240 ± 94.51 mg·kg^{-1}), and station 3 (20 220 ± 193.13 mg·kg^{-1}) belongs also to this class, while the recorded values at the sites 4 with *CI* of 9.05 (18 100 ± 246.5 mg·kg^{-1}), and station 5 with *CI* of 4.66 (9320 ± 541.27 mg·kg^{-1}) belong to class 2 [ABRMC 1991]. Hence, their levels exceed the inferior limits in the major parts of the study zone, excepting the station 6 which doesn't exhibit iron pollution. Here, a decreasing concentration gradient of iron was remarkably seen from station 2 to station 6 (Figs. 2, 3). The relative important values of copper are recorded at the three stations: station named Sidi Salem (St1), having only depth of 10 m, along with average concentration about of 81.9 ± 8.17 mg·kg^{-1} and *CI* of 3.15, Station of Wadi Seybouse (St2) presenting an average concentration of 86.32 ± 7.6 mg·kg^{-1} and *CI* of 3.32, and the last station is located in the harbour (St4), belonging to class 2 with the following average values of 104 ± 10.15 mg·kg^{-1} and *CI* of 4 [ABRMC 1991] (Figs. 2, 3). Furthermore, our findings showed a clear variability of lead (Pb) level from one site to another, it varies from the maximum of 129.58 ± 9.73 mg·kg^{-1} and *CI* of 5.89 in the harbour station (St4) belonged to class 2, and the minimum of 21.53 ± 3.08 mg·kg^{-1} and *CI* of 0.97 in Ain-Achir beach (St6). Also, we noticed that *CI* of 5.29 (116.38 ± 16.6 mg·kg^{-1}) and 4.94 (108.68 ± 19.58 mg·kg^{-1}) recorded in station of Wadi Seybouse and the station of Sidi Salem are also part of class 2 [ABRMC 1991] (Figs. 2, 3). Regarding zinc concentration, the sediments of the whole study zone belong to class 1, excepting station 3 that belongs to class 2, and found as a non-negligible contamination site with *CI* value of 3.34 (293.92 ± 9.91 mg·kg^{-1}). The cadmium levels show that the sediment of the west coastal of Annaba Gulf belongs to class 1 with a maximum value of *CI* about 2.5 (1.503 ± 0.186 mg·kg^{-1}) at the harbour (St4). The relative results of nickel levels in the superficial sediments of the western region of Annaba Gulf showed variability in metal levels that do not explain metal pollution [ABRMC 1991], and hence maximum values of 67.95 mg·kg^{-1} and *CI* of 1.51 (St4) were noticed [ABRMC 1991] (Figs. 2, 3). The ANOVA test revealed a very highly significant spatial difference ($p \leq 0.001$) for all the studied metals in superficial sediments. Consequently, the quantitative distribution of the metal levels in sediments of Annaba Gulf follows this order: Fe > Pb > Cu > Zn > Ni > Cd.

The results of pollution load index (*PLI*) (10 m) and *PLI*s (20 m) of various elements are given in Figure 4. The range of *PLI*s results were of 0.65 to 3.22, and reaching the maximum value at the station 4 and the minimum one at the station 6 of 10 m of depth. The *PLI* of stations 1, 2, 3 and 4 (only for a depth of 10 m) are up than 1 (Fig. 4). Thus, according to total levels of *PLI*s, the classification order of different stations in term of enrichment is as follows: St4 (Harbour) > St2 (Wadi Seybouse) > St1 (Sidi Salem) > St3 (Joinoville) > St5 (Rizi Amor) > St6 (Ain-Achir). The obtained results of ANOVA test has shown a very highly significant difference ($p \leq 0.001$) between the sampling sites. The Figure 4 shows the variation of *PLI*s at different places.

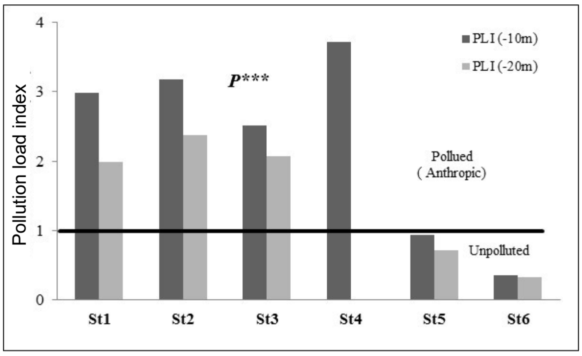

Fig. 4. Variations of the pollution load index (*PLI*) in the sediment samples collected from various stations; St1–St6 as in Fig. 2; source: own study

According to the French guidelines as well as a special view point, stations 1, 2 and in particular station 4, a showing a marked polymetallic contamination by Fe and Pb, in addition to a slight contamination by Cd, making the sediment of these three stations a polluted zone of the highest importance on the Algerian coast. This is the most urbanized part of the bay, receiving the main exits of waste waters, whether 13 sewers discharging all forms of wastes (urban, industrials and pluvial). Thus, the network between the bay and Seybouse River surrounding by an important siege of industrial activity, containing various industries, and consequently the domestic and industrial wastes are discharged in the river. Nevertheless, the generalized contamination of sediments of Annaba Gulf confirms the existence of a pollution source. Indeed, the coastal waters receive indirectly by watershed of Seybouse River, containing the industrial wastes coming from steel alloy manufacture of El Hadjar (10 km South of Annaba Gulf known as the highest industrial pole and the most diversified industry in Africa [BELABED *et al.* 2017], as well as they are directly affected by port/export of iron-ore of Annaba. Hence, the highest levels of zinc, lead and copper are noticed in the harbour, explaining the involved effect of the port activities and the large number of emissaries of the domestic waste.

This could be due to the wide use of antifouling paint in hull boat coverings [AUGIER *et al*. 1992], and to road traffic emissions containing high levels of lead (Pb), and hence the metal contamination can be whether through the direct atmospheric disposition or the indirect way following the washout of road dust

Table 4. Concentration of heavy metals in the sediment of Annaba Gulf and other coastal ecosystems in the whole world

Area	Zn	Cu	Pb	Fe	Cd	Ni	References
	mg·kg^{-1} dry weight			%	mg·kg^{-1} dry weight		
West coast of Annaba Gulf	2.72–293.92	13.22–104	21.5–108.68	2.4–226.8	0.001–1.5	0.76–67.95	present study
Average Continental Crust	52	25	14.8	–	0.1	–	WEDEPOHL 1995
Annaba Gulf (Algeria)	190.2–301.1	15.0–60.3	10.04–186.1	13.1–459.91	0.9–2.62	17.3–51.6	BELABED *et al.* 2013b
Ghazaouet Gulf (Algeria)	190.24	39.53	31.78	14.50	1.68	–	BENGUEDDA-RAHAL 2012
Gabs Gulf (Tunisia)	5.2–716.5	0.59–5.8	3.8–13.9	–	0.11–950	–	EL ZRELLI *et al.* 2015
Atlantic Coast (Morocco)	378.98	98	159.98	195.17	0.003–0.07	–	NADEM *et al.* 2015
Aghien Coast (Ivore coast)	10.68–55.11	59.98–243	0.16–3.41	0.002–0.133	0.15	–	TRAORE 2014
Gulf of Guinea	2.6–82.2	0.2–29.3	21–22.2	–	0.1–0.4	1.4–66.75	MAHU *et al.* 2015
Jezan Coast (Saudi Arabia)	24.74	16.39	3.86	–	0.48	14.32	GOLAM-MORTUZA *et al.* 2017
Toulon Harbour (France)	15–1880	5.8–1080	14–710	–	0.004–3.4	8–66	TESSIER 2012
San Pietro Island (Italy)	35–62	8.05–22.34	14.28–29.19	–	0.2–1.7	9–36.17	DI LEO *et al.* 2013
Andam Island	10.4–27.27	6.64–7.04	–	0.508–3.93	0.69–1.96	2.16–2.88	NOBI *et al.* 2010
South–East coast of India	39.73–72.68	1.35–15.75	11.85–23.05	1.75–4.35	0.59–6.41	39.1–59.76	BARATH *et al.* 2017
Coastal Pakistan	26.68–111.3	0.14–77.41	24.68–42.39	0.96–1.02	1.11–1.46	43.03–51.31	SAHER, SIDDIQUI 2016
Bengal Bay (Bangladesh)	–	6.1	1.42	–	0.79	7.25	KHAN *et al.* 2017
Laizhou Bay (China)	38.22–73.81	7.57–21.29	9.65–17.65	–	0.11–0.28	12.85–25.35	ZHANG, GAO 2015
Baixada Santista (Brazil)	5.81–133.64	0.70–30.73	1.29–48.19	1.53–43.79	–	0.94–19.27	KIM *et al.* 2016

Source: own elaboration.

by rainwater [CONOR 1980]. The regulatory presence of nickel and cadmium could be related to domestic waste effluents and urban sewers (fuels, batteries and other electrical apparatus, as well as the mud bottom of harbour is considered to be as a carrier substrate on which nickel can be highly absorbed [GUILLON-COTTARD 1997]. The significant concentration with metal detected at a depth of 10 m. Indeed, the collected sediments from the coastal region are very close to waste zones, exhibiting that all elements present at concentrations slightly very higher from the coast to the depth. This result is in line with those obtained by [CHRISTOPHORIDIS *et al.* 2009] in the Gulf of Thermai (Greece), and by [MC ALISTER *et al.* 2005]. These authors have found that metals entering into waters and sediments are transported and enter the geochemical cycle and are slightly retained at the coastal edges.

The pollution load index (*PLI*) of the west coastal sediments of Annaba Gulf was calculated in order to understand the changes in the contamination levels of sediments in various stations. Very low values of *PLI* (<1) do not involve appreciable anthropic effects [TOMLINSON *et al.* 1980]. Also, the calculated values of *PLI* showed that the studied sediments, excepting those of stations St5 and St6 were up than 1. This enrichment indicated that these sediments were polluted by heavy metals resulting from the common or different original source (*PLI* > 1), dumping urban and industrial waste effluents in these stations. Additionally, the selected metals in this study are considered as an original crust of stations 5 and 6, where *PLI* becomes inferior than 1 (*PLI* < 1).

The comparison of trace element levels with those reported for another coastal region in the average continental crust [WEDEPOHL 1995] are given in Table 3. In the study zone, the average concentrations of Pb, Cd, Zn and Cu have showed higher values than those found in the average continental crust [WEDEPOHL 1995] (Tab. 4). The current values of iron are higher than those of the other coastal regions, whilst the reported levels for the Annaba Gulf [BELABED *et al.* 2013] were of the same order of magnitude than those of the present study. Moreover, the obtained results of nickel are comparable to different reports for other coastal regions, since the concentration of zinc is comparable or superior to those of other regions of Tunisia and France. In addition, the higher concentrations of Zn, Cu and Pb were found as 15–1880 mg·kg^{-1}, and 14–710 mg·kg^{-1} respectively, in the Toulon Harbour of France [TESSIER 2012] (Tab. 4).

METALLIC LEVELS IN FISH TISSUES

Figure 5 shows a strong variability between the sampling stations, indicating that the higher levels are those of iron in all harvests, with a maximal value of 199.67 ± 17.5 mg·kg^{-1} recorded in the harvested individuals at the station of Wadi Seybouse, and a minimal value of 95.87 ± 11.5 mg·kg^{-1} recorded in the harvested individuals at the station of Ain-Achir. Unlike to zinc concentration in stations 5 and 6, zinc exhibits higher values than those of the recommended nutritional guidelines given by WHO/USEPA, with a maximum value of 161 ± 1.82 mg·kg^{-1} recorded in the harvested fishes in station 4 (Fig. 5). Moreover, the results of fishes caught from the studied stations, excepting those of station 6, have shown high values of lead, exceeding the guideline values [EC 2006], and subsequent they reach the maximal value for the individual lots of station 4 (2.39 ± 0.092 mg·kg^{-1}). On the other hand, the concentration of the remaining

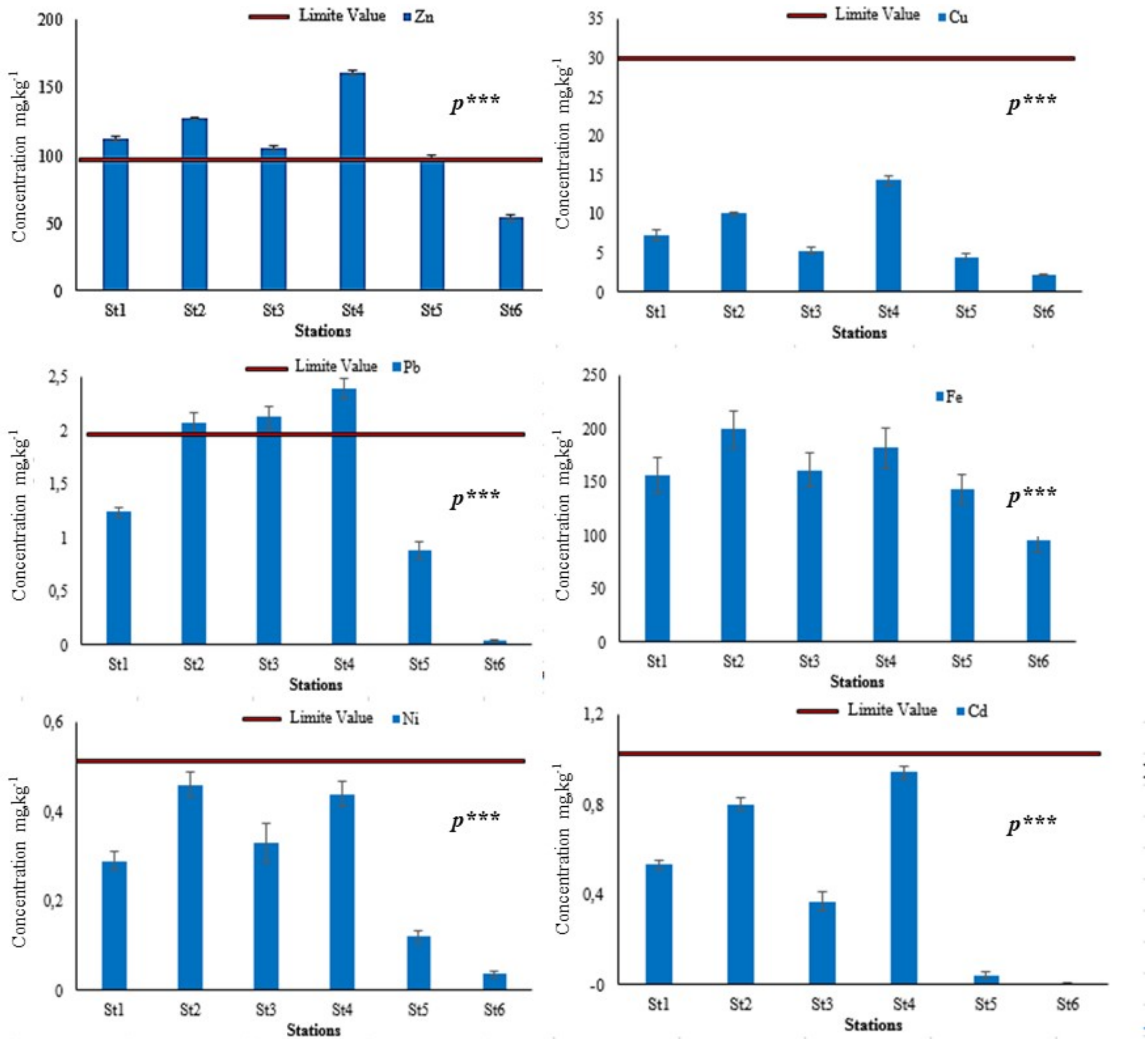

Fig. 5. Mean concentrations of metals in fish tissues (mg·kg^{-1} dry weight) of the six selected stations; $p \leq 0.001$; source: own study

metals (Cu, Ni and Cd) in fish muscles of all harvest are lower than the maximal value limits given by WHO/USEPA, and therefore the maximum values were found to be as 14.35 ± 0.61 mg·kg^{-1}, 0.46 ± 0.027 mg·kg^{-1} and 0.8 ± 0.042 mg·kg^{-1} for copper, nickel and cadmium, respectively (Fig. 5). In contrast, the cadmium level is 0.05 mg·kg^{-1}, as given by the regulations of European Community [EC 2006], and therefore the caught fishes in stations 1, 2, 3 and 4 are contaminated by metals (Tab. 3). The data analyses showed the following bioaccumulate order of heavy metals in *M. cephalus* issue: Fe > Zn > Pb > Cu > Ni > Cd. Similarly of the sediment, the ANOVA test revealed a very highly significant spatial difference ($p \leq 0.001$) for all the studied metals in muscle of fish.

The enrichment order of the individuals in stations for the metallic elements obeys to their concentration order in sediments. Noteworthy, the higher levels of metals were recorded in fish samples collected from the proximity of human activities, such as beaches of harbor, Sidi Salem, Joinoville, and station of Wadi Seybouse. This metal enrichment is strongly related to the industrial wastes of steel complex conveyed by Seybouse River to Gulf, and to the road axis, as well as harbour activity leading to metal contaminations, including lead, copper, zinc, and in particular iron. The lead is used as an anti-detonator in gasoline, since the contamination by this element is related to anthropic activities [LI *et al.* 2003]. The agriculture activity could also promote the increase of copper, zinc and cadmium concentrations. As far as known, *M. cephalus* is a pelagic, omnivorous species, and narrowly related to the sediments, and also it's characterized by weak mobility and food rich by algae, polychaetes, crustaceans, gastropods and fishes [STANCHEVA *et al.* 2013]. The sediment forms a reservoir of metals and the other aquatic environmental pollutants. In relation to their mobility and food preferences [FOWLER 1986], the fishes survive close to the sea bottom, and thus reflect the concentrations of the environmental metallic elements [ENNOURI *et al.* 2013]. Importantly, the bioaccumulation is also related with feeding, swimming and the metabolic activity of the individual and species, and may depend on size,

Table 5. Comparison of metal concentrations in muscle of flathead grey mullet (*Mugil cephalus*) of coastal region of Annaba with those found in others in the world (mg·kg^{-1} dry wt)

Area	Zn	Cu	Pb	Fe	Cd	Ni	Reference
West coast of Annaba Gulf	107.98	8.10	1.21	147.73	0.475	0.248	present study
FAO/WHO [a)]	100	30	2	–	2	1	1984
SEPA [b)]	100	20	9	–	10	0.2	2005
CE	–	–	0.30	–	0.050	–	2006
Middle East coast (Tunisia)	180	19.12	–	–	0.28	–	HAMZA-CHAFFAI *et al.* [1996]
North-East Mediterranean (Turkey)	37.39	4.41	5.32	–	0.66	–	CANLI and ATLI [2003]
Black Sea (Turkey)	86.2	2.14	0.68	–	0.35	–	YILMAZ [2009]
Lake Köyceğiz – Mugla (Turkey)	394.4	25.4	1.72	–	0.48	–	
Coast of Rio de Janeiro (Brazil)	0.02	0.005	0.0008	–	0.0001	–	MEDEIROS *et al.* [2012]
Lake Bafa; Eastern Aegean (Greece)	14.9	1.25	0.91	–	0.0235	–	AYDIN-ONEN *et al.* [2015]
Coast of Dakar (Senegal)	160	373	0.39	–	1.79	–	DIOP *et al.* [2016b]
Estuary of Saint Louis (Senegal)	98	180	0.08	–	2.31	–	

[a)] Codex Alimentarius Commission 1984. [b)] SEPA China 2005.
Source: own study and own elaboration acc. to literature.

Table 6. Pearson correlation coefficient analysis of trace metals in fish and the coastal sediment of Annaba

Variables	Zn1	Zn2	Zn 3	Cu 1	Cu 2	Cu 3	Pb 1	Pb 2	Pb 3	Fe 1	Fe 2	Fe 3	Ni 1	Ni 2	Ni 3	Cd 1	Cd 2	Cd 3
Zn1	1																	
Zn2	0.997***	1																
Zn 3	0.899**	08.23*	1															
Cu 1	0.976***	0.983***	0.867**	1														
Cu 2	0.987***	0.984***	0.858**	0.992***	1													
Cu 3	0.888**	0.803*	0.952***	0.861**	0.889**	1												
Pb 1	0.903**	0.873**	0.946***	0.937***	0.921**	0.908**	1											
Pb 2	0.916**	0.923**	0.852**	0.974***	0.965***	0.918**	0.960***	1										
Pb 3	0.941***	0.918**	0.885**	0.889**	0.893**	0.799*	0.852***	0.768*	1									
Fe 1	0.828*	0.965***	0.759*	0.886**	0.967***	0.644	0.884***	0.943**	0.856**	1								
Fe 2	0.965***	0.974***	0.918**	0.964***	0.969***	0.851**	0.942***	0.937**	0.918**	0.999***	1							
Fe 3	0.842**	0.840**	0.888**	0.845**	0.885**	0.812*	0.918**	0.843**	0.911**	0.897**	0.898**	1						
Ni 1	0.878**	0.861**	0.944***	0.905**	0.890**	0.857**	0.986***	0.911**	0.873**	0.912**	0.950***	0.933***	1					
Ni 2	0.809*	0.825**	0.966***	0.853**	0.859**	0.888**	0.981***	0.905**	0.776*	0.940***	0.926***	0.895**	0.994***	1				
Ni 3	0.957***	0.953***	0.886**	0.957***	0.979***	0.864**	0.929***	0.921**	0.947**	0.900**	0.946***	0.946***	0.911**	0.844**	1			
Cd 1	0.941***	0.914***	0.915**	0.966***	0.948***	0.927***	0.962***	0.995**	0.817**	0.807*	0.938***	0.807*	0.920**	0.918**	0.900**	1		
Cd 2	0.963***	0.901**	0.962***	0.909**	0.922**	0.985***	0.923**	0.866**	0.992***	0.791*	0.903**	0.917**	0.895**	0.854**	0.962***	0.904**	1	
Cd 3	0.940***	0.887**	0.904**	0.950***	0.953**	0.954***	0.942***	0.961***	0.844**	0.782*	0.882**	0.867**	0.884**	0.834**	0.949***	0.958***	0.961***	1

Explanations: 1 = 10 m of depth, 2 = 20 m of depth, 3 = in fish muscle, * = $p \le 0.05$, ** = $p \le 0.01$, *** = $p \le 0.001$.
Source: own study.

feeding habits, ability of biological concentration of every species [KWOK *et al.* 2014]. This study, therefore presents new information about the distribution of metals in fishes along of Annaba Gulf.

It's interesting to compare the concentrations found in muscles of *Mugil cephalus* collected from Annaba Gulf with those obtained in other regions in the world subjected also to anthropic pressure (Tab. 5).

Among all the metal elements of the studied zone, only zinc concentrations do not conform with the dietary guidelines, and thus pose a human health risk according to FAO/WHO (Tab. 5). Regarding zinc and copper concentrations, the most studies on various regions of the world revealed consistent concentrations with dietary guidelines, reaching a high concentration of 394.4 mg·kg^{-1} for zinc, and 180 mg·kg^{-1} for copper [DIOP *et al.* 2016]. Overall, the mullet collected from Senegal was found to have higher muscular concentrations for copper, cadmium and zinc compared to dietary guidelines.

The Pearson correlation coefficient analysis and the significance levels of the obtained results are given in Table 6, indicating that all coefficients are strongly correlated ($p \le 0.5$, 0.01, 0.001), and consequently these heavy metals are likely resulted from the common source: natural or anthropic. Besides, a weak correlation (0.644) was found between copper and iron (Tab. 6). The significant positive correlation calculated using Pearson correlation coefficient test analysis confirms the existence of homogeneity between the presence of metals in sediments and fish.

CONCLUSIONS

The results showed that the sediments of Annaba Gulf contain some of the studied metals. The determination of *CI* and *PLI* revealed an important polymetallic contamination by iron, lead and copper in stations 1, 2, 3 and 4, and however, slightly important for cadmium where the most metal concentrations become slightly inferior at the depth (20 m). Addionally, the concentrations of heavy metals in muscles of mullet (*M. cephalus*) were also lower than those of the current world guidelines (WHO/USEPA, EC), except zinc, cadmium and lead concentrations in stations 1, 2, 3 and 4. On the other part, the positive correlation was observed between the metal concentrations in sediments and tissues of *M. cephalus,* indicating that these elements are released from common origin of the anthropic activity. These metallic element enrichments are somehow related to domestic and industrial wastewater inputs of the town that flow in these stations, without pretreatment process. Indeed, zinc, copper and lead are considered as metallic elements, characterizing the urban pollution and hence the continued monitoring would be mainly carried out in the polluted zones, in particular, the port station in which important metallic waste discharges, including the studied metals were remarkably noticed. Moreover, the high levels and hazardous of zinc and lead bioaccumulations in *M. cephalus* fish of Annaba Gulf are of concerns for the authorities, due to the tendency of these metals in causing serious body diseases. A regular monitoring program for heavy metals is recommended in order to protect these compartments and to reduce the environmental risks. The most protected places are beach of Rizi Amor, and especially station of Ain-Achir. The later does not receive any waste discharges, and thereby it benefits a continuing renewal of waters following the current regimen of the bay (high currentology).

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Ocena zanieczyszczenia metalami osadów i tkanek cefala pospolitego (*Mugil cephalus* Linnaeus, 1758) ze wschodniego wybrzeża Algierii

STRESZCZENIE

W pracy oceniono poziom zanieczyszczenia strefy przybrzeżnej w okolicach Annaby na podstawie spektrofotometrycznych analiz metali (Fe, Cu, Pb, Zn, Ni and Cd) w osadach na różnej głębokości (10 i 20 m) i w mięśniach cefala pospolitego (*Mugil cephalus*) jako organizmu wskaźnikowego. W okresie zimowym (2014 r.) zebrano 12 próbek osadów powierzchniowych i łącznie 24 ryby. Po wysuszeniu, pokruszeniu i przesianiu próbek do stężonych kwasów dodano 0,5 g suchej masy każdej próbki. Wyniki dowodzą, że stężenie niektórych metali przekracza dopuszczalne normy, a osady w zatoce są zanieczyszczone żelazem, ołowiem i miedzią. Wskaźnik zanieczyszczenia (*CI*) kształtował się następująco: Fe > Pb > Cu > Zn > Ni > Cd, przy czym większe stężenie większości metali notowano na głębokości 20 m.

Średnie stężenie metali w mięśniach ryb układało się natomiast w porządku: Fe > Zn > Pb > Cu > Ni > Cd. Konsumpcja ryb z niektórych skażonych stanowisk może być niebezpieczna, ponieważ stężenie ołowiu, kadmu i cynku przekracza międzynarodowe standardy. Ponadto, silna dodatnia korelacja między stężeniem metali w osadach i w rybach dowodzi, że metale te pochodzą z tych samych antropogenicznych źródeł takich jak zrzuty ścieków bytowych, portowych, przemysłowych i rolniczych. Potwierdza to oznaczony wskaźnik ładunku zanieczyszczeń (*PLI*). W podsumowaniu zaleca się wdrożenie regularnego programu monitoringu w celu ochrony organizmów wodnych i ograniczenia ryzyka środowiskowego.

Słowa kluczowe: *głębokość wody, metale ciężkie, osad, ryby, toksyczność*