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Cadmium, copper, lead, and zinc in *Mugil cephalus* from seven coastal lagoons of NW Mexico

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Abstract The increasing order of the mean concentrations of Cd, Cu, and Zn in the tissues of *Mugil cephalus* of seven coastal lagoons of Sinaloa State (NW Mexico) was liver > gills > muscle, while for Pb it was gills > muscle ≥ liver. There were no differences between the mean concentrations of Cd and Pb of the three tissues determined in the samples of the seven lagoons and, although there were some significant differences, there was no indication of a latitude-related trend in the distribution of Cu and Zn: the Cu content of the muscle tended to be higher in the northern than in the southern lagoons, although in the case of the gills the highest and lowest mean values indicated an opposite trend, with the highest and lowest values in one southern and one northern lagoon. In the case of the liver, there were no differences and no indication of a regional trend. There were no differences in the mean Zn contents of muscle and gills; in the case of the liver, one of the lagoons of the central part of the state had a significantly higher value than one of the southern lagoons and all the rest had similar values. In addition, there was no clear indication of season-related differences in any of the three tissues. According to our results, the metal contents of the muscle of this species are not of concern for human health, since the allowable ingestion would be in the order of 0.9 kg/day.

Keywords Lagoons · Metal pollution · Fish consumption · NW Mexico

Introduction

Metal pollution of the marine and coastal environment is a worldwide concern, because of their toxicity and accumulative behavior. For this reason, monitoring of heavy metals is essential to assess their potential threat to the environment, as well as to human health. The levels of heavy metals are

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increasing in the coastal lagoons and salt marshes of the Mexican NW, in part because of human activities, mainly intensive agriculture and food-related industry (Soto-Jiménez et al. 2003), as well as because of the geomorphological, physical, and chemical characteristics of the alluvial soils of the Mexican Pacific coastal plains (Frías-Espericueta et al. 2006). This is of concern, since these water bodies are important ecosystems because of their high biodiversity and complexity, high productivity and marked variety of habitats suitable as breeding and nursery areas for many aquatic organisms.

In addition, there has been a growing interest to assess the levels of heavy metal contamination of fish and other fishery products since, in view of their high contents of high quality protein and polyunsaturated fatty acids, they are considered to support good health. However, their consumption may be a significant pathway to metal exposure in the human population living in coastal areas, because these organisms accumulate metals from the aquatic environment (Türkmen et al. 2008; Yildirim et al. 2009).

For this reason, several fish species have been used in marine/coastal pollution monitoring programs, because they are good indicators of metal accumulation in the marine environment (Çoğun et al. 2006; Dural et al. 2006). In coastal lagoons, mullets have been used in several of these studies because, being filter and detritus feeders (Drake et al. 1984; Minos et al. 1995), they are exposed to the contaminants present in water and sediments (Blasco et al. 1998; Fernandes et al. 2007, 2008). For this reason, they have been suggested as good candidates for pollution monitoring studies (Ferreira et al. 2004; Bu-olayan and Thomas 2005).

There is limited information on the heavy metal contents of fish from NW Mexico and, since the grey mullet *Mugil cephalus* is widely exploited and consumed in the region, we evaluated the possible risk of its consumption determining the Cd, Cu, Pb, and Zn contents of the edible muscle, as well as of the gills and liver of commercial landings of this species in seven coastal lagoons of Sinaloa state (NW Mexico).

Material and methods

Study area

The lagoons of Navachiste (NAV), Santa María–La Reforma (SMLR), Altata–Ensenada del Pabellón (AEP), Ceuta (CE), Urías (UR), Huizache–Caimanero (HUC) and Teacapán–Agua Brava (TAB; Fig. 1) support traditional fisheries and the main anthropogenic impacts are the effluents from intensive agriculture and rural communities, as well as from semi-intensive shrimp culture systems. Additional impacts are the municipal and industrial wastewaters of the cities of Culiacán and Navolato (≈950,000 inhabitants) in the case of AEP, and of the city of Mazatlán (≈400,000 inhabitants), of its harbor and of several food-processing industries in the case of UR.

Sampling

The fish samples (15 organisms in each case) were obtained from local fishermen of each lagoon, during the months of March, May, July, September to November 2006 (depending on road conditions and fish availability), and January 2007.

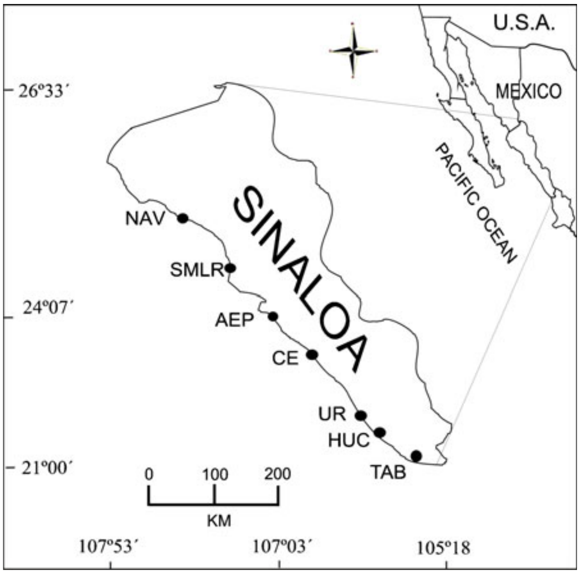

Fig. 1 Geographic position of the seven lagoons

Since age and size may be important factors in metal accumulation (Türkmen et al. 2008), only adults of similar size were collected. After sampling, the organisms of each sample were transported to the laboratory in metal-free separate plastic bags kept in ice boxes.

Chemical analysis

All the organisms of each sample were dissected and approximately 100 g of muscle, and the entire gills and liver were separated. The sample of each tissue was freeze-dried, ground and homogenized in a Teflon mortar. After this operation, triplicate 1 g subsamples, spiked with known amounts of the four metals (Miller and Miller 1988), were digested in 25 ml concentrated HNO_3 (trace metal analysis), evaporated slowly to dryness (90°C) and the solid residue was redissolved in 2M HNO_3. Finally, the samples were centrifuged and the supernatant was used for metal analysis by flame atomic absorption spectrophotometry.

The purity of reagents and the presence of possible contaminants were determined with one blank for each set of five to six samples, using the same procedure used for the samples (Türkmen et al. 2009). Deionized (MilliQ) water was used throughout the study. All materials used for sample handling and for metal analysis were acid-washed (Moody and Lindstrom 1977), and the accuracy of the method was assessed using DOLT-4 dogfish certified reference material (National Research Council Canada): the percentages of recovery ranged from 91.5% to 105.6%. The metal concentrations, corrected for these percentages, are reported on a dry weight basis.

Statistical analysis

Since the data were not normal (Lilliefors' test), the differences in the mean metal contents of each tissue (calculated using the individual values of the 35 samples of each tissue obtained in the seven lagoons) were determined using one-way repeated measures nonparametric ANOVA tests.

To determine differences between lagoons, the mean metal contents found in the five samples of each tissue of each lagoon were compared by one-way ANOVAs. When the data were not normal or homoscedastic (Lilliefors' and Bartlett's tests), the data were compared with the equivalent nonparametric tests and the different means were separated with Tukey's or Dunn's tests, for parametric and nonparametric ANOVAs, respectively. In all cases, the level of significance was $\alpha = 0.05$.

Results and discussion

Comparisons between tissues

The mean concentrations of the three tissues indicate different capacities of metal accumulation. The increasing order of concentration for Cd, Cu, and Zn was: liver > gills > muscle, and the concentrations in the muscle were significantly lower than those of liver and gills, whereas for Pb the order was gills > muscle ≤ liver (Table 1). In all cases, the data confirm that the muscle is not actively involved in metal accumulation (Kargin and Erdem 1991; Erdoğrul and Erbilir 2007).

It has been shown that the liver is highly active in metal storage as well as in metal detoxification since, as a response to metal exposure, metallothionein induction occurs mainly in fish liver (Roesijadi and Robinson 1994; Tepe et al. 2008), which explains the high concentrations of Cd, Cu, and Zn found in this tissue.

The high Pb levels observed in the gills coincide with the results of Çoğun et al. (2006), who found that Pb concentrations were higher in the gills

Table 1 Mean concentrations (μg/g, dry weight) of Cd, Cu, Pb, and Zn in the muscle, gills, and liver of *Mugil cephalus* of seven lagoons of Sinaloa (NW Mexico)

	Muscle	Gills	Liver
Cd	0.17 ± 0.10a	0.66 ± 0.13b	2.03 ± 1.05c
Cu	1.06 ± 0.56a	4.63 ± 1.94b	305.21 ± 160.17c
Pb	1.63 ± 1.21a	16.68 ± 13.70b	1.43 ± 1.24a
Zn	21.77 ± 5.48a	86.96 ± 8.09b	167.00 ± 69.19c

Different letters indicate significant differences between tissues (one-way repeated measures nonparametric ANOVA tests, $\alpha = 0.05$; a < b < c) of *M. cephalus* than in other tissues, probably through sorption and Pb complexation in the mucus of the gill lamellae, followed by desorption and active uptake in the gill microenvironment (Tao et al. 1999).

Comparisons between lagoons

The mean Cd concentrations in the muscle ranged from 0.23 ± 0.18 to 0.13 ± 0.09 μg/g, and the respective ranges for gills and liver were 0.75 ± 0.22 to 0.59 ± 0.07, and 2.47 ± 1.37 to 1.34 ± 1.28. There were no significant differences between lagoons, although the values tended to be higher in UR, TAB, and NAV, and lower in SMLR (Table 2). Since all these lagoons are close to agricultural areas, agricultural effluents are likely to be the main Cd sources, because this metal is associated to the minerals used to manufacture phosphate fertilizers (Alloway 1995; Lugon-Moulin et al. 2006).

Cu concentrations ranged from 430.1 ± 157.2 to 204.8 ± 80.8 in liver, and the respective ranges in gills and muscle were between 7.04 ± 2.91 and 3.47 ± 1.95, and from 1.72 ± 0.73 to 0.62 ± 0.17 μg/g, respectively. There were no differences between the mean values determined in the fish livers of the seven lagoons, whereas the mean concentration of the gills was significantly lower in NAV than in HUC, and the remaining lagoons had intermediate values. In the case of the muscle, the highest mean value was that found in SMLR, followed by that determined in NAV, which were significantly higher than those of UR and TAB (Table 2).

In the muscle and liver, Pb concentrations ranged from 2.10 ± 0.98 to 1.24 ± 1.47 μg/g, and between 2.26 ± 1.57 and 0.71 ± 0.96 μg/g, respectively. The gills had higher values, ranging from 30.60 ± 21.5 to 7.84 ± 1.33 μg/g and, as in the case of Cd, there were no significant differences between the mean values of the seven lagoons (Table 2).

Mexico is one of the leading Pb producers, and this metal is present in most mineral ores of interest to the mining industry. Due to its widespread

Table 2 Annual mean metal concentrations (μg/g, dry weight) in the tissues of *Mugil cephalus* of Sinaloa State (NW Mexico)

Lagoon	Cd	Cu	Pb	Zn
Muscle				
NAV	0.23 ± 0.18a	1.47 ± 0.58bc	1.95 ± 1.28a	23.00 ± 4.22a
SMLR	0.13 ± 0.09a	1.72 ± 0.73c	1.66 ± 1.02a	24.77 ± 8.65a
AEP	0.14 ± 0.12a	1.31 ± 0.22abc	2.10 ± 0.98a	22.33 ± 3.54a
CE	0.17 ± 0.10a	0.86 ± 0.44ab	1.53 ± 1.41a	19.13 ± 3.89a
UR	0.16 ± 0.05a	0.62 ± 0.28a	1.24 ± 1.47a	21.55 ± 5.92a
HUC	0.16 ± 0.07a	0.84 ± 0.14ab	1.38 ± 1.39a	20.80 ± 8.14a
TAB	0.17 ± 0.10a	0.62 ± 0.17a	1.58 ± 1.41a	20.82 ± 3.08a
Gills				
NAV	0.64 ± 0.06a	3.47 ± 1.95a	7.84 ± 1.33a	84.47 ± 3.71a
SMLR	0.59 ± 0.07a	3.77 ± 0.96ab	8.02 ± 1.95a	85.48 ± 6.06a
AEP	0.62 ± 0.08a	4.94 ± 2.32ab	15.03 ± 9.56a	92.66 ± 5.93a
CE	0.65 ± 0.18a	4.23 ± 0.68ab	26.67 ± 17.50a	80.83 ± 7.61a
UR	0.69 ± 0.16a	5.07 ± 0.99ab	17.00 ± 10.05a	86.45 ± 5.72a
HUC	0.69 ± 0.11a	7.04 ± 2.91b	30.60 ± 21.54a	92.57 ± 14.30a
TAB	0.75 ± 0.22a	3.90 ± 0.89ab	11.60 ± 5.78a	86.27 ± 6.10a
Liver				
NAV	2.46 ± 1.07a	204.81 ± 80.85a	1.32 ± 1.31a	179.16 ± 50.65ab
SMLR	1.34 ± 1.28a	285.57 ± 244.55a	0.71 ± 0.96a	144.23 ± 33.99ab
AEP	1.72 ± 0.88a	394.77 ± 68.17a	0.82 ± 1.14a	228.72 ± 61.26b
CE	2.22 ± 0.99a	430.01 ± 157.22a	2.26 ± 1.57a	184.60 ± 67.78ab
UR	2.47 ± 1.37a	368.92 ± 215.92a	1.68 ± 1.36a	180.35 ± 88.89ab
HUC	2.36 ± 0.52a	248.07 ± 84.36a	1.80 ± 1.35a	76.20 ± 9.92a
TAB	1.62 ± 1.05a	204.34 ± 64.55a	1.41 ± 0.87a	175.76 ± 63.33ab

Equal or common letters indicate lack of significant difference between the mean content of each metal determined in the same tissue of different lagoons (one-way ANOVA tests, $\alpha = 0.05$; $a \leq ab \leq abc$; $a < b < c$). Pb (gills): nonparametric test use and because of its abundance, it is one of the metals of concern for public and environmental health in Mexico (Flores and Albert 2004). It is released into the environment from natural and anthropogenic sources and, though it might have accumulated in previous years in lagoon sediments from Pb-added gasoline emissions, other likely important sources are the contributions from the mineral-rich soils of the Sierra Madre Occidental. Whatever its origin, the high Pb values (>80 to >140 μg/g) found in the sediments of AEP lagoon (Green-Ruiz and Páez-Osuna 2003), are probably matched in other lagoons, which would explain the results of this study, because of the feeding habits of *M. cephalus*.

In the case of Zn, the only significant difference was between the highest and lowest mean values found in the liver samples of HUC (76.20 ± 9.92 μg/g) and of AEP (228.72 ± 61.26 μg/g), whereas there were no differences between lagoons in muscle and gills (19.13 ± 3.89 to 24.77 ± 8.65 and 80.83 ± 7.61 to 92.66 ± 5.93 μg/g, respectively: Table 2).

The state of Sinaloa has 11 rivers draining into its coastal waters and lagoons, which flow through the metal-rich soils of the Sierra Madre Occidental, and receive the surplus waters used for irrigation of the >600,000 ha of agricultural land, mostly dedicated to intensive, highly mechanized cultures.

Additional sources of metals are the effluents of industrial, municipal, aquaculture, mining and harbor activities, all of which contribute to the economy of the State of Sinaloa (Anonymous 2004). Most of these effluents drain directly or indirectly into estuarine and lagoonal systems where the levels of contaminants tend to be high, especially in their inner parts, because of the long flushing times and low renovation rates of their waters (Montaño-Ley et al. 2008).

There are several references to season-related variations of metal concentrations, which are generally related to the seasonality of some human activities such as agriculture (Burger et al. 2005; Çoğun et al. 2006; Dural et al. 2007). In NW Mexico, the rainy season is from July to October, when the increase in the volumes of water drained into the lagoonal systems was expected to be reflected in higher metal contents of our samples. However, the only significant differences were the

Table 3 Metals concentrations (μg/g, dry weight) in tissues of Mugilidae from different regions

Species	Site	Tissue	Cd	Cu	Pb	Zn
Liza saliens[a]	Paramos lagoon, Portugal	Liver	–	262 ± 140	<DL	88.6 ± 32.0
		Muscle	–	<2.6	<DL	25.7 ± 10.1
Mugil cephalus[b]	Aegean Sea	Muscle	0.45 ± 0.03	1.26 ± 0.10	0.61 ± 0.04	40.2 ± 3.3
Mugil sp.[c,d]	Fayoum, Egypt	Liver	1.45	13.7	13.9	24.7
		Muscle	1.10	5.92	13.3	18.3
Mugil cephalus[e]	Tuzla Lagoon, Turkey	Liver	0.02–0.35	4.7–12.03	1.85–3.12	26.7–49.7
		Muscle	0.08–0.11	0.47–0.62	0.49–1.19	8.27–60.8
Mugil cephalus[f]	Camlik lagoon Turkey	Liver	0.94			113.2
		Muscle	0.06			101.1
Mugil cephalus[g]	Northeast Mediterranean	Liver	5.9–8.1	79.6–98.6	11.2–16.8	69.8–86.5
		Muscle	1.1–2.2	8.3–12.9	5.7–7.8	21.5–29.7
Mugil cephalus[h]	NW Mexico	Liver	1.34–2.47	204–430	0.71–2.26	76–228.7
		Muscle	0.13–0.23	0.62–1.72	1.24–2.10	19.1–24.8

<DL: below detection limits.

[a] Fernandes et al. (2008)

[b] Uluozlu et al. (2007)

[c] Mansour and Sidky (2002)

[d] Converted to dry weight from the original data

[e] Dural et al. (2007)

[f] Dural et al. (2006)

[g] Çoğun et al. (2006)

[h] This study

higher values of Pb found in the liver during the rainy season (1.990 ± 1.235 μg/g), in comparison to the 1.056 ± 1.125 μg/g of the dry season, and the 90.39 ± 9.524 μg/g of Zn determined in the rainy season, which was significantly higher than the 84.670 ± 6.207 μg/g of the dry season.

On the other hand, contradictory to the result found in the case of the liver, the Pb content of the muscle was 1.974 ± 1.218 μg/g in the dry season, significantly higher than in the rainy season (1.120 ± 1.029 μg/g). In addition, the Cd contents of muscle and gills were significantly higher in the dry (0.208 ± 0.095 and 0.717 ± 0.131 μg/g) than in the rainy season (0.101 ± 0.083 and 0.582 ± 0.094 μg/g).

In view of these results, it appears that the variability of the metal contents of *M. cephalus* of the seven lagoons object of this study is not related to meteorological events, but is more probably due to the continuous high volumes of the effluents draining into rivers and lagoonal systems, which are required by the intensive agricultural practices of Sinaloa State.

Table 3 compares the results of this study to those reported for mugilidae in other lagoonal and marine ecosystems. In the case of the liver, the mean contents of the four metals are among the higher values available in the literature, whereas the concentrations found in the muscle are in the low part of the range reported worldwide.

When regularly ingested, toxic substances can be harmful even at low concentrations. For this reason, fish muscle is commonly analyzed to determine if its consumption might pose a health risk. The reference values mentioned in USFDA (1993) and WHO (1998) were compared to the range of annual mean values calculated for each lagoon, showing that the consumption of this species does not represent a serious concern for human health, since the minimum allowable daily ingestion would be 0.9 kg/day of *M. cephalus* muscle (Table 4).

Table 4 Provisional tolerable daily intake (PTDI) (in μg/person/day) of Cd, Cu, Pb and Zn (USFDA 1993; WHO 1998) and range of allowable limits of daily ingestion (grams, wet weight/person/day) of edible muscle of *M. cephalus*

Metal	PTDI	grams/day
Cd	55	991–1,685
Cu	3,000	7,137–19,749
Pb	750	1,300–2,468
Zn	45,000	7,414–9,667

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