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Differential Tissue Accumulation of Copper, Iron, and Zinc in Bycatch Fish from the Mexican Pacific

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Abstract In order to ascertain if Cu, Fe, and Zn are differentially accumulated in fish tissues, metal concentrations were measured in the muscle and liver of bycatch fish from the states of Sinaloa (189 specimens, 7 species) and Guerrero (152 individuals, 8 species) in the Mexican Pacific Coast during March and November 2011. Additionally, metal levels were compared with the maximum allowable limits set by international legislation and contrasted with similar ichthyofauna from other regions. Liver had more elevated concentrations of Cu (Sinaloa 28.3, Guerrero 16.3 μg g^{-1}), Fe (Sinaloa 1098, Guerrero 636 μg g^{-1}), and Zn (Sinaloa 226, Guerrero 186 μg g^{-1}) than the muscle in fish from both studied areas. The relative abundances of analyzed metals in both tissues was Fe > Zn > Cu. As far as limits set by international legislation (Australia, India, New Zealand, Zambia), measured concentrations of Cu in the edible portion of fish were not found to be above the set values. In the case of Zn, the maximum allowable limits set by international legislation were exceeded by the Peruvian mojarra *Diapterus peruvianus* from Guerrero state (Mexican Pacific). No limits exist for Fe in the edible portion of fishery products in the national and international legislations.

Keywords Bycatch fish · Essential metals · Muscle and liver · Mexican Pacific

Introduction

A broad classification of metals divides them as essential, beneficial, or detrimental. Trace elements recognized as essential for biota, including human health, include iron, zinc, copper, chromium, iodine, cobalt, molybdenum, and selenium [1] as they are involved in important biological processes [2]. These elements play a functional and structural role in the human body [3]. Fish is a major source of iron in adults and children and a deficiency of it causes anemia. Fish is significant to the human diet due to its high protein content, low saturated fats, and abundant omega fatty acids known to support good health. On the other hand, trace metals can pose a risk to fish and humans, especially elements of elevated toxicity and facility to be biomagnified [4]. In this context, various studies on contamination of different fish species by trace metals have been conducted worldwide; in the specific case of bycatch fish, few studies have been carried out in Mexican waters [5, 6]. In the Mexican Pacific coast, shrimp fishery is an important source of bycatch resources, which include fish, crustaceans, and mollusks [7]. In terms of abundance, fish makes up the most sizeable group as it has been estimated that for every kilogram of shrimp, 10 kg of fish are caught [8, 9]. Fish muscle tissue is the most frequently used for analysis because it is a major target tissue for metal storage and it constitutes the main edible part of the fish [10].

Trace metals in the aquatic environment can affect aquatic biota and fish consumers, such as humans and other wildlife. These metals may enter the coastal ecosystem from different natural and anthropogenic sources, including industrial, aquaculture, agriculture, and domestic sewage, besides atmospheric

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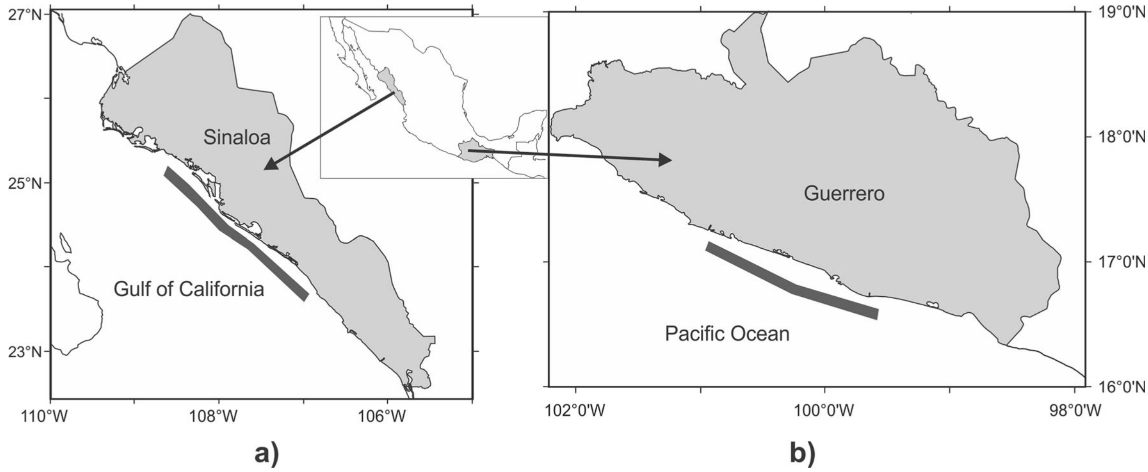

Fig. 1 Location of shrimp trawling areas where bycatch fish were obtained. **a** Sinaloa State. **b** Guerrero State

precipitation [11–13]. The utilization of phosphorus-containing products such as fertilizers and detergents has also contributed to an increase of trace metals in water bodies. In aquatic ecosystems, trace metals have received considerable attention due to their toxicity and accumulation in fishes. The state of Sinaloa has a littoral of 656 km and an area of coastal lagoons of 221,600 ha [14]. Such coastal waters are of great ecological and economic relevance; the climate is sub-tropical-humid (AW type), with rainfall in summer and less than 10 °C of annual thermal oscillation [15]. In Mexico, the coast of Sinaloa is home to the highest concentration of aquatic farms, especially for shrimp production [16]; nevertheless, the intensive agriculture practiced in the surroundings of the state releases great quantities of agrochemicals [17], including fertilizers and fungicides containing metals. The coast of Guerrero has approximately 600 km of coastline, an amount that places it as the eighth longest state in the country, plus an area of 70,000 ha of inland waters and lagoons. In most of the state territory (82 %) the climate is warm and humid; the rest of the surface has dry, semi-dry, and temperate humid climate: the average annual temperature is 25 °C, the average minimum temperature is 18 °C, and maximum 32 °C; most of the rainfall

Table 1 Feeding habits and concentrations (minimum, maximum, average, and standard deviation) of copper, iron, and zinc ($\mu g\ g^{-1}$ dry weight) in the muscle and liver of the fish collected in Sinaloa

Common name	Scientific name	FH	N	Tissue	Cu	Fe	Zn
Peruvian mojarra	*Diapterus peruvianus*	C	57	M	0.65–5.66 ($1.34^{a,b}$ ± 0.76)	24.9–330 ($86.1^{a,c}$ ± 50.1)	30.1–77.9 ($44.7^{b,c}$ ± 10.3)
				L	2.45–35.4 ($10.18^{a,b}$ ± 6.61)	231–6944 ($1544^{a,c}$ ± 1399)	0.65–5.66 ($202^{b,c}$ ± 122)
Toothed flounder	*Cyclopsetta querna*	O	8	M	0.56–1.39 ($0.95^{a,b}$ ± 0.28)	25.4–143 (72.7^{a} ± 49.8)	15.4–39.8 (24.5^{b} ± 8.93)
				L	5.19–25.9 (11.3^{a} ± 6.71)	410–1238 (620^{a} ± 278)	86.4–265 (155 ± 56.3)
Silver weakfish	*Isopisthus remifer*	O	12	M	0.75–1.2 ($0.97^{a,b}$ ± 0.12)	15.7–136 (49.0^{a} ± 31.8)	25.6–74.3 (48.2^{b} ± 15.8)
				L	0.69–17.9 ($7.96^{a,b}$ ± 4.91)	161.6–2632 (947^{a} ± 854)	63.0–631 (298^{b} ± 151)
Silver drum	*Larimus argenteus*	O	37	M	0.59–1.31 ($0.92^{a,b}$ ± 0.17)	27.6–112 ($56.5^{a,c}$ ± 18.4)	21.5–61.4 ($35.2^{b,c}$ ± 8.92)
				L	1.59–181 ($28.8^{a,b}$ ± 28.4)	110–11,746 ($1886^{a,c}$ ± 2444)	11.6–526 ($170^{b,c}$ ± 92.3)
Yellowstripe grunt	*Haemulopsis axillaris*	C	46	M	0.73–2.48 ($1.29^{a,b}$ ± 0.35)	19.6–116 (43.6^{a} ± 19.7)	18.0–63.0 (33.7^{b} ± 10.6)
				L	4.7–579 ($46.6^{a,b}$ ± 90.6)	305–3093 ($1272^{a,c}$ ± 619)	71.4–350 ($196^{b,c}$ ± 62.8)
Slender croaker	*Micropogonias ectenes*	C	17	M	1.18–2.36 ($1.60^{a,b}$ ± 0.30)	26.8–180 ($44.2^{a,c}$ ± 35.6)	16.0–26.8 ($20.3^{b,c}$ ± 3.10)
				L	20.4–162 ($64.0^{a,b}$ ± 42.2)	311–2144 ($758^{a,c}$ ± 464)	20.4–162.2 ($64.0^{b,c}$ ± 42.2)
Blackblotch pompano	*Trachinotus kennedyi*	C	12	M	1.30–2.71 ($1.76^{a,b}$ ± 0.43)	23.6–94.4 (43.5^{a} ± 19.4)	19.1–69.5 (33.7^{b} ± 14.3)
				L	11.6–77.1 ($29.5^{a,b}$ ± 18.9)	218–1121 (660^{a} ± 285)	117–3938 (495^{b} ± 1085)
Average				M	1.24 ± 0.36	56.5 ± 16.7	34.3 ± 9.96
				L	28.3 ± 21.0	1098 ± 484	226 ± 138

FH feeding habits, *C* carnivore, *O* omnivore, *N* number of individuals, *M* muscle, *L* liver

For a given tissue and fish species, same superscript letters indicate significant ($p < 0.05$) differences occurs in summer, June through September; the average rainfall in the state is 1200 mm annually [18]. The aims of this study were as follows: (a) to determine Cu, Fe, and Zn distribution in the muscle and liver of bycath fish from selected sites of the Mexican Pacific coast; (b) to compare levels of Cu, Fe, and Zn with maximum permissible limits in fishery products set in the international legislation; and (c) to contrast metal levels in analyzed ichthyofauna with similar species worldwide.

Materials and Methods

Fish samplings were conducted in the Mexican Pacific; in the state of Sinaloa, 189 specimens of 7 species were collected; in Guerrero, the number of individuals was 152 and they belong to 8 species. According to a national survey on environmental impacts in Mexican coastal cities, in Sinaloa coasts, the factors that mostly affect the aquatic environment are tourism and recreational activities and habitat destruction; such activities may contribute to the occurrence of trace metals. In Guerrero state, the main environmental impacts are represented by wastewater discharges, tourism, and recreation and habitat destruction [19].

Bycatch fish were collected during shrimp trawling operations in the continental shelf of the states of Sinaloa (Fig. 1a) and Guerrero (Fig. 1b) according to their fishing plans. Fish from Sinaloa were obtained during March 2011 and fish samples from Guerrero were collected in November 2011, at a depth range of 30–46 m. In the laboratory, fish were identified and total length and weight recorded. Muscle tissue from the median dorsal portion and the liver were used for analysis. Samples were frozen at −20 °C and lyophilized for 72 h (−52 °C and 60 × 10^{-3} mbar) in a Labconco Freeze-dry-System-FreeZone 6; then, they were ground in an agate mortar

Table 2 Feeding habits and concentrations (minimum, maximum, average and standard deviation) of copper, iron and zinc (μg g^{-1} dry weight) in the muscle and liver of the fish collected in Guerrero

Common name	Scientific name	FH	N	Tissue	Cu	Fe	Zn
Bigscale goatfish	*Pseudupeneus grandisquamis*	C	13	M	0.64–2.83 ($1.27^{a,b}$ ± 0.59)	18.6–90.1 (29.0^{a} ± 19.1)	18.5–65.9 (30.4^{b} ± 12.2)
				L	5.94–27.5 ($12.3^{a,b}$ ± 5.87)	238–2148 ($588^{a,c}$ ± 488)	118–312 ($190^{b,c}$ ± 56.0)
Yellowfin jack	*Hemicaranx leucurus*	O	10	M	0.84–5.57 ($3.08^{a,b}$ ± 1.47)	26.6–56.1 (42.1^{a} ± 9.31)	16.6–36.6 (24.0^{b} ± 6.78)
				L	4.18–10.2 ($7.54^{a,b}$ ± 1.74)	192–599 (356^{a} ± 116)	88.0–266 (171^{b} ± 46.6)
Three-spot flounder	*Ancylopsetta dendritica*	C	5	M	0.46–1.19 ($0.82^{a,b}$ ± 0.29)	14.2–46.9 (26.8^{a} ± 13.9)	17.6–48.6 (28.7^{b} ± 12.2)
				L	4.41–54.1 (21.2^{a} ± 19.6)	157–1088 (544^{a} ± 388)	82.2–195.8 (141 ± 49.0)
Peruvian mojarra	*Diapterus peruvianus*	C	49	M	0.28–6.17 ($1.43^{a,b}$ ± 1.01)	19.0–2801 (107^{a} ± 395)	17.9–357.5 (49.3^{b} ± 47.2)
				L	5.09–28.0 ($10.4^{a,b}$ ± 4.46)	337 ± 3254 ($945^{a,c}$ ± 559)	43.4–298.3 ($168^{b,c}$ ± 72.8)
Yellowstripe grunt	*Haemulopsis axillaris*	C	24	M	1.02–3.88 ($1.94^{a,b}$ ± 0.82)	23.7–170 (51.1^{a} ± 31.4)	16.6–66.1 (34.4^{b} ± 12.9)
				L	7.16–107 ($31.3^{a,b}$ ± 28.7)	186–2024 ($867^{a,c}$ ± 452)	94.6–239 ($167^{b,c}$ ± 39.9)
Slender croaker	*Micropogonias ectenes*	C	32	M	1.09–3.02 ($1.78^{a,b}$ ± 0.49)	24.4–239 (52.1^{a} ± 45.1)	18.0 ± 69.7 (34.7^{b} ± 17.1)
				L	6.39–65.1 ($19.0^{a,b}$ ± 13.6)	216–2389 ($882^{a,c}$ ± 412)	104–1032 ($225^{b,c}$ ± 204)
Silver drum	*Larimus argenteus*	O	10	M	0.18–1.97 ($1.44^{a,b}$ ± 0.49)	21.4–93.7 (37.3^{a} ± 21.0)	16.1–34.6 (28.3^{b} ± 5.7)
				L	11.5–32.4 ($19.2^{a,b}$ ± 6.81)	248–847 ($543^{a,c}$-201)	80.8–169 ($128^{b,c}$ ± 28.3)
Blackblotch pompano	*Trachinotus kennedyi*	C	9	M	0.99–7.26 ($4.38^{a,b}$ ± 2.10)	26.4–205 (64.9^{a} ± 54.4)	8.61–78.3 (26.6^{b} ± 20.3
				L	2.39–26.9 ($9.33^{a,b}$ ± 7.04)	90.6–734 (366^{a} ± 194)	104–676 (299^{b} ± 159)
Average				M	2.01 ± 1.16	51.3 ± 25.8	32.0 ± 7.85
				L	16.3 ± 7.91	636 ± 233	186 ± 54.4

FH feeding habits, *C* carnivore, *O* omnivore, *N* number of individuals, *M* muscle, L liver

For a given tissue and fish species, same superscript letters indicate significant ($p < 0.05$) differences

Table 3 Maximum permissible limits (μg g^{-1} wet weight) of Cu and Zn in fresh fish for human consumption and maximum values (μg g^{-1} wet weight) in muscle of fish in this study

Element	Legislation	Permissible limits[a]	Species	Maximum value[b]	Site
Cu	Australia	10	*Trachinotus kennedyi*	2.03	Guerrero
	India	10	*Diapterus peruvianus*	1.58	Sinaloa
	New Zealand	30			
	Zambia	100			
Zn	Australia	40	*Diapterus peruvianus*	100.1	Guerrero
	India	50	*Diapterus peruvianus*	21.8	Sinaloa
	New Zealand	40			
	Zambia	100			

[a] According to [27], no limits have been set for Fe

[b] concentrations were converted from dry weight to wet weight considering 72 % of humidity

with pestle (Fisher-Scientific). Powdered samples (0.25 g) were acid digested in duplicates (5 mL of concentrated nitric acid-trace metal grade, Baker) using capped Teflon vials (Savillex™) on a hot plate (Barnstead Thermolyne) during 3 h (120 °C). Digested samples were stored in polyethylene containers for further analysis. For metal analysis, an atomic absorption spectrophotometer (Varian SpectrAA220) was used. For Zn and Fe, flame atomic absorption spectrophotometry was used (FAAS); the working conditions were as follows: wavelengths (nm) 248.3 (Fe) and 213.9 (Zn) and lamp current was 10 mA for both elements. In the case of Cu, measurements were made by graphite furnace atomic absorption spectrophotometry (GF-AAS); the working conditions were wavelength 324.7 nm and temperature for sample atomization was 2100 °C. The accuracy and precision of metal measurements were ensured using blanks and certified reference material for trace metals (liver DOLT-4, NRC-Canada). Elemental recoveries were estimated as the ratio of measured concentrations with respect to the corresponding value in the certificate of the reference material. Measured concentrations (n = 8) of Fe (1650 μg g^{-1}), Cu (33.7 μg g^{-1}), and Zn (127.5 μg g^{-1}) in the reference material were comparable to certified levels (Fe 1833 μg g^{-1}, Cu 31.2 μg g^{-1}, and Zn 116 μg g^{-1}). Recovery percentages were appropriate (Fe 90 %, Cu 108 %, Zn 110 %). The limits of detection of Cu, Fe, and Zn were 0.01, 0.19, and 0.48 μg g^{-1}, respectively. Metal concentrations are reported in microgram per gram on a dry weight basis; for comparisons with legal limits of metals in fishery products (reported as μg g^{-1} on a wet weight basis) conversions of concentrations from dry weight to wet weight were made considering an average humidity of 72 % in the muscle tissue of analyzed fish. Comparisons of elemental concentrations in a given tissue were made by a one-way analysis of variance (ANOVA-Kruskal-Wallis test); in the case of

Table 4 Concentrations (μg g^{-1} dry weight; mean ± standard deviation) of Cu, Fe, and Zn in the edible part of Mullidae and Carangidae fish

Species	Cu	Fe	Zn	Site	Reference
		Mullidae			
Mullus surmuletus	0.80	NA	6.24	Andalusia, Spain	[28]
Pseudupeneus grandisquamis	1.27 ± 0.59	29.01 ± 19.12	30.41 ± 12.19	Guerrero, Mexico	This study
		Carangidae			
Trachurus trachurus	1.70 ± 0.63	14.95 ± 2.50	38.39 ± 4.59	Granada, Spain	[29]
Trachurus trachurus	5.85	NA	46.97	Andalusia, Spain	[28]
Trachurus trachurus	0.15 ± 0.01	18.16 ± 0.14	5.49 ± 0.01	Edo State, Nigeria	[30]
Trachurus mediterraneus	1.95 ± 0.73	23.55 ± 20.10	35.52 ± 11.40	Granada, Spain	[29]
Trachurus picturatus	5.96 ± 4.18	48.41 ± 9.76	77.76 ± 48.05	Granada, Spain	[29]
Trachinotus kennedyi	1.76 ± 0.43	43.46 ± 19.38	33.66 ± 14.26	Sinaloa, Mexico	This study
Trachinotus kennedyi	4.38 ± 2.1	64.94 ± 54.43	26.63 ± 20.29	Guerrero, Mexico	This study

NA not available

significant differences, a Dunn's test was used. A $p < 0.05$ level was considered of statistical significance. All statistical analyses were performed using GraphPad Prism 4.0.

Result and Discussion

The main contributors of metals in fish are food and surrounding water, with diet being the dominant source [20]. When elements are ingested, they are distributed all over the fish body and bound to diverse sites in tissues and organs, which implies that metals are distributed differentially. For the three analyzed elements, liver showed more elevated concentrations than muscle. The highest metal concentrations were measured in the liver of fish from both areas. As far as relative abundances of analyzed elements, the sequence of average concentrations in muscle and liver of fish from both areas was Fe > Zn > Cu. Metal concentrations in the muscle and liver of fish species from Sinaloa are presented in Table 1; the highest mean Cu concentration was found in the liver of *Micropogonias ectenes* (64.0 μg g^{-1}) and the lowest concentration was present in the muscle of *Larimus argenteus* (0.92 μg g^{-1}). In the case of Fe, the highest value was measured in the liver of *L. argenteus* (1886 μg g^{-1}) while the lowest was found in the muscle of *Trachinotus kennedyi* (43.5 μg g^{-1}). For Zn the highest level was detected in the liver of *T. kennedyi* (495.2 μg g^{-1}); the smallest concentration was found in the muscle of *M. ectenes* (20.3 μg g^{-1}). In fish from Guerrero state, the highest average Cu concentration was found in the liver of *Haemulopsis axillaris* (31.3 μg g^{-1}); the lowest value (0.82 μg g^{-1}) was in *A. dendritica* muscle (Table 2). In the case of Fe, the highest value was detected in the liver of *Diapterus peruvianus* (945 μg g^{-1}) while the lowest was found in the muscle of *Ancylopsetta dendritica* (26.8 μg g^{-1}). For Zn, the highest level was measured in *T. kennedyi* liver (299 μg g^{-1}); the lowest concentration was found in the muscle of *Hemicaranx leucurus* (24.0 μg g^{-1}). At low levels, metals like Cu, Fe, and Zn are essential for enzymatic activity and diverse biological processes in fish [21]; although essential elements are required for fish metabolism, they may become toxic at elevated levels. Some of the toxic effects of essential metals on fish include the alteration of physiological activities and biochemical parameters in blood [22]. Metals such as Cu, Fe, and Zn are essential since they play important structural and functional roles in biological systems [23]. In this context, Fe is mostly found in hemoglobin and hemosiderin in fish liver [24]. On the other hand, it has been found that Zn can accumulate less in fish muscle, whereas fish liver is able to store Cu in higher amounts compared to muscle and gills [25] because this organ works as a Cu deposit [26]. Mexican legislation does not set maximum permissible levels of Cu, Fe, and Zn in fishery products, so legal limits of Cu and Zn from other countries were used as guideline values (Table 3). Limits for Fe in the international legislations do not exist. For comparative reasons, only the species with the highest values of Cu and Zn in muscle tissue were included; it can be seen that legal limits of Cu were not exceeded. With regard to Zn, *D. peruvianus* from Guerrero exceeded all the maximum permissible limits in legislation from all the areas studied [27]. Taking into account that the Peruvian mojarra *D. peruvianus* is widely consumed in Mexico and that the level of Zn in this species from both sampling areas exceeded the legal limits, it is necessary to monitor Zn availability through *D. peruvianus*. In order to contrast our results with studies from other areas, levels of Cu, Fe, and Zn were compared among similar fish species of the families Mullidae and Carangidae (Table 4). In Mullidae, only a study with *Mullus surmuletus* from Spain was published [28]; levels of Cu and Zn were an order of magnitude higher in our study. As far as Carangidae, the reported studies matched three species of *Trachurus*. Levels of Cu, Fe and Zn in *T. kennedyi* from Sinaloa and Guerrero (this study) were comparable to concentrations reported in fish from Spain [28, 29]. In contrast, Cu and Zn concentrations in our study were higher than reported values in *Trachurus trachurus* from Nigeria. It is worth mentioning that samples from the Edo State in Nigeria [30] were purchased in local markets as smoked fish, which implies that samples might have been caught in different areas. Moreover, the thermal process may alter the elemental composition as compared to raw tissue so conclusions should be made with caution.

Concluding Remarks In general, bioaccumulation of Cu, Fe, and Zn in fish from both areas followed the order liver > muscle. The average concentrations of the analyzed elements in both tissues was Fe > Zn > Cu. With respect to the maximum permissible limits in fishery products for human consumption, Cu concentrations in the edible portion of fish were not exceeded; Zn levels were above those allowed by international legislation [27] in the muscle of the Peruvian mojarra *D. peruvianus* from Guerrero. Concentrations of Fe in studied fish were not compared because no legal limits exist in the national and international legislations. The studied fish belong to families Mullidae and Carangidae; in the case of Mullidae, our fish had Cu and Zn levels higher than *M. surmuletus* from Spain [28]. Regarding Carangidae, our fish had higher levels of Cu and Zn than similar fish from Nigeria [30] but comparable to levels reported in fish from Spain.

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Compliance with Ethical Standards

Conflict of Interest The authors declare that they have no conflict of interest.

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