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Health Risks Associated with Heavy Metals in Imported Fish in a Coastal City in Colombia

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Abstract

Colombia is a fish exporter and consumer country because of its geographical location. Since 2012, imported fish have become a more economical option than domestic species due to free trade agreements. Concentrations of Pb, Cd, and Zn were evaluated in three imported and highly commercialized fish species in a city on the Caribbean coast of Colombia: *Prochilodus lineatus*, *Prochilodus reticulatus*, and *Pangasianodon hypophthalmus*, plus one brand of canned tuna and one brand of sardines. The canned species showed the highest values for Pb, Cd, and Zn; canned tuna (oil-packed) contained 0.189 ± 0.047 mg/kg Pb and 238.93 ± 76.43 mg/kg Zn, while canned sardines contained 0.111 ± 0.099 mg/kg Cd, suggesting a relationship between the canning process and the metal concentrations. The estimated daily intake (EDI) and hazard quotient (HQ) suggested that there is no risk for consumer health in the short term, but the presence of these heavy metals certainly should be a concern in the long term because of the bioaccumulation phenomenon due to the high intake of these fish species in this coastal and tourist community. It is recommended that continuous monitoring of heavy metal concentrations take place to protect communities in a local and global context.

Keywords Heavy metals · Imported fish · Canned tuna · Potential risk · Colombia

Introduction

Fish are an important source of easily digested proteins that contain essential amino acids and are a good source of polyunsaturated fatty acids, particularly omega-3 fatty acids, which are known to reduce the risk of cardiovascular disease and help the nervous system [1]. Nevertheless, fish and shellfish pose a health risk, and their consumption becomes hazardous due to the contamination of the environment in which they grow and live [2]. In many bodies of water where fishing activities occur, there is also heavy metal contamination as the result of anthropogenic mining and industrial activities [3, 4]. Due to heavy metal toxicity, high persistence, biomagnification, bioaccumulation, and nonbiodegradability in food chains, heavy metals constitute a group of contaminants that generate the most negative impacts on aquatic ecosystems [5–7].

Heavy metals such as zinc (Zn) are essential for fish and human metabolism, while others, such as cadmium (Cd) and lead (Pb), have no known function in biological systems. For normal fish metabolism, essential metals come from water, food, and sediments. However, similar to

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essential metals, nonessential metals are also absorbed by and bioaccumulated in fish tissues depending on age, gender, exposure period, trophic level, and fish dietary habits [8–13].

It has been established that for different fish species, bioaccumulation and biomagnification occur at different trophic levels. The water column and bottom sediments represent a potential health risk for humans [6, 14–17]. Heavy metals in fish are a problem that has already been addressed [18–21] but still elicits great interest because of the possible medium- and long-term consequences for organisms that consume them, including humans. There are no reports about this issue in fish species imported and consumed in the coastal city of Barranquilla and introduced to Colombia through this city.

The international trade in fish and fishing products has grown in recent decades due to free trade agreements and the increase in aquaculture activity, among other aspects [2]. According to the Food and Agriculture Organization (FAO), fresh fish are the most important fishery product, followed by frozen and canned fish. One third of the world's fish production is traded internationally. Therefore, aspects such as product quality and consumer safety guarantees have become important to distributors [2]. Since 2012, Colombia, despite being a producer country in the global fishing industry, has been facing a situation in which imported fishery products (fresh, frozen, and canned) have lower prices compared to native products because of its entry into FTAs (free trade agreements). Three decades ago, the consumption of fish in Colombia was 3.7 kg/year; today, the per capita consumption of fishery products is 4.73 kg/year. However, this is considered low compared to other fish producers, such as Japan (54 kg/year) and Spain (38 kg/year). It is also low when compared with the average consumption in Latin America (18 kg/year). According to data reported by the National Administrative Department of Statistics of Colombia (DANE), imports in the fishing and aquaculture sector reported 1.8% growth between 2015 and 2016 [22]. Approximately 198,579 tons of fish and shellfish consumed in the country come from imports, and the remaining quantity comes from domestic production, meaning that 77.48% of consumed fish is imported, which is why this investigation is relevant.

Since 2013, imports have come mainly from China, Vietnam, Ecuador, Chile, Senegal, South Africa, and the USA [23]. The most consumed imported fish species by the local market are the basa fillet (*P. hypophthalmus*) and canned tuna (oil-packed and water-packed). Native species are not always available throughout the year; therefore, easily accessible and inexpensive imported fish have become a popular food of choice because they are readily available. The species selected for this investigation come from locations where there has been confirmation of contamination by heavy metals [24–27]. The main objective of this study is to determine the content of heavy metals (Pb, Cd, and Zn) in samples of fish species that were fresh, frozen, and canned and imported and marketed in the city of Barranquilla, Colombia. Additionally, this study seeks to calculate the human health risk of regular intake of these species, which is an important information for consumer health.

Materials and Methods

Sample Collection and Analysis

Samples of canned tuna, canned sardines, and three fish species (fresh and frozen) were selected in local stores and supermarkets located in Barranquilla, a city located on the Caribbean coast of Colombia, from September 2016 to March 2017. For each fish species and canned fish sample, 20 specimens and 20 cans were obtained, respectively, totaling 100 samples. Fresh fish samples were "bocachico" from Argentina *(P. lineatus)* and "bocachico" from Venezuela *(P. reticulatus)*, while frozen fish samples were basa fillet *(P. hypophthalmus)* from Vietnam. Canned sardines in tomato sauce were from Ecuador, and canned tuna (oil-packed and water-packed) was from China. All the species were duly identified, the places of origin were declared by the supplier, and the homogeneity of the size of the samples of fresh and frozen fish was taken into account. The canned fish samples were selected according to the brands of tuna and sardines that are certified by Colombia's National Institute of Food and Drug Surveillance's (INVIMA) health registry.

Samples of muscle tissue in each fresh and frozen specimen were extracted according to the procedure described in the Reference Methods for Marine Pollution Studies, RSRM 11 [28]. The sample extraction was performed by cutting and removing a 3-cm wide (10 g) portion of skin; the muscle sample was placed in a polyethylene bag previously washed with a 5% HNO_3 solution and deionized water for its preservation and analysis. For the canned specimens, 10 g was taken from the content of each can, drained, isolated, and immediately frozen at − 20 °C; frozen samples were stored until heavy metals were determined.

The heavy metal analysis (Pb, Cd, and Zn) was performed by atomic absorption spectrophotometry using a graphite furnace (Thermo Scientific™ iCE™ series 3500) after acid digestion of the sample (0.5 g *w/w*) with HNO_3/HCl (3:1 *v/v*) for 3 h at 95 °C [16]. Analytical quality control was performed using certified reference material IAEA 407, and the recovery rate ranged between 91% and 98%. The detection limits for the different metals were 0.010 μg/g for Cd, 0.011 μg/g for Pb, and 0.016 μg/g for Zn. All reagents were of analytical grade from Merck Millipore.

Human Health Risk Assessment

To evaluate the health risk associated with heavy metal contamination for imported fish, the estimated dietary intake (EDI), the hazard quotients (HQ), the maximum allowable fish consumption rate (CR_{lim}), and the metal pollution index (MPI) were estimated in this study [16, 29]. A simple food survey was conducted with inhabitants in the same locations where the samples of imported fish were taken. A total of 150 people from the general population were randomly selected: 30 of them were children (2–10 years old), 68 were women, and 52 were men (20–58 years old). The participants were asked about several aspects related to imported fish consumption habits (i.e., the total amount and type of imported fish consumed per week and meal size). The sampling population was divided into two groups: children (CHD) and the rest of the population, including women and men (RP).

The potential human health risk assessment was conducted by considering the estimated daily intake (EDI) (μg/kg bw/day), calculated using the following equation:

$$EDI = \frac{Cm\ x\ DI}{BW} \tag{1}$$

where Cm is the mean metal concentration in edible muscle tissue (μg/g), DI is the intake of every fish consumed per day (g/day), and BW is the mean body weight of the participants. In the present study, 35 kg for CHD and 70 kg for RP were assumed. According to the diet survey ($n = 150$), the following mean values of daily fish intake for canned tuna (0.041 kg), canned sardines (0.046 kg), basa fillet (0.048 kg), *P. lineatus* (0.044 kg), and *P. reticulatus* (0.055 kg) were used to calculate the EDI.

Risk may be characterized using a hazard quotient (HQ). This is the ratio of the EDI of a chemical to a reference dose (RfD) (mg/kg/day) defined as the maximum tolerable daily intake of a specific metal that does not result in any deleterious health effects:

$$HQ = \frac{EDI(mg/kg)}{Reference\ Dose\ (Rfd)(mg/kg/day)} \tag{2}$$

There would be no obvious hazard if the value of HQ was less than 1. If HQ > 1, then the EDI of a particular metal exceeds the RfD, indicating that there is a potential risk associated with that metal.

According to the US EPA [30], the maximum allowable fish consumption rate (CR_{lim}, g/day) of contaminated fish considering the noncarcinogenic effect of a contaminant can be calculated as follows:

$$CRlim = \frac{RfD\ x\ BW}{Cm} \tag{3}$$

The metal pollution index (MPI) was computed to determine the overall heavy metal concentration detected in fish and canned fish. This index was obtained by calculating the geometrical mean of the concentrations of all the analyzed metals [31]. The heavy metals used for computing the MPI were Pb, Cd, and Zn.

$$MPI(\mu g/g) = (Cf_1 x\ Cf_2 x \ldots Cf_n)^{1/n} \tag{4}$$

where Cf_n is the metal concentration in the sample.

Statistical Analysis

The results of the analysis for each species are presented as the mean ± the standard deviation of the analyzed samples. Concentration values were checked for normality by using the Kolmogorov–Smirnov test, and there was no normality in the data. The differences between fish species and canned fish were tested for significance using the nonparametric Student's *t* test and Kruskal–Wallis test, establishing a significance level of $p \leq 0.05$ to determine significant differences among the heavy metal concentrations in fish species. In addition, Pearson's linear correlation analysis was used to establish the association among the heavy metal concentrations, and the results again show that there was no correlation. All statistical analyses were performed with SSPS 18 (PASW Statistics 18).

Results and Discussion

Figure 1 for Pb, Fig. 2 for Cd, and Fig. 3 for Zn show the heavy metal concentrations detected in the muscle tissue of all samples of the studied fish species. Among these metals, Zn exhibited the highest mean concentration in all the canned species. The Zn concentration was 236.6 mg/kg for water-packed tuna, 238.9 mg/kg for oil-packed tuna, and 141.6 mg/kg for canned sardines; these values are double, and even triple, the values of other species. Oil-packed tuna also reported the highest mean concentration of Pb (0.19 mg/kg), while the highest levels of Cd were reported in canned sardines (0.11 mg/kg); values that are above the maximum limits of the FAO/WHO [29], the European Union (EU) [32], and Colombia for heavy metals in fish [33] (Table 1). On the other hand, canned tuna (oil-packed and water-packed) had the lowest values for Cd (0.03 mg/kg) in all species. The detritivore species *Prochilodus* (bocachicos) and the omnivorous species *P. hypophthalmus* (basa fillet) have similar patterns in the concentrations of Zn and Cd. It was observed that the canned species, despite being at opposite ends of the trophic chain as carnivores (tuna) and filter feeders (sardines), both reported high values for Pb and Zn, suggesting perhaps a linkage to the canning process.

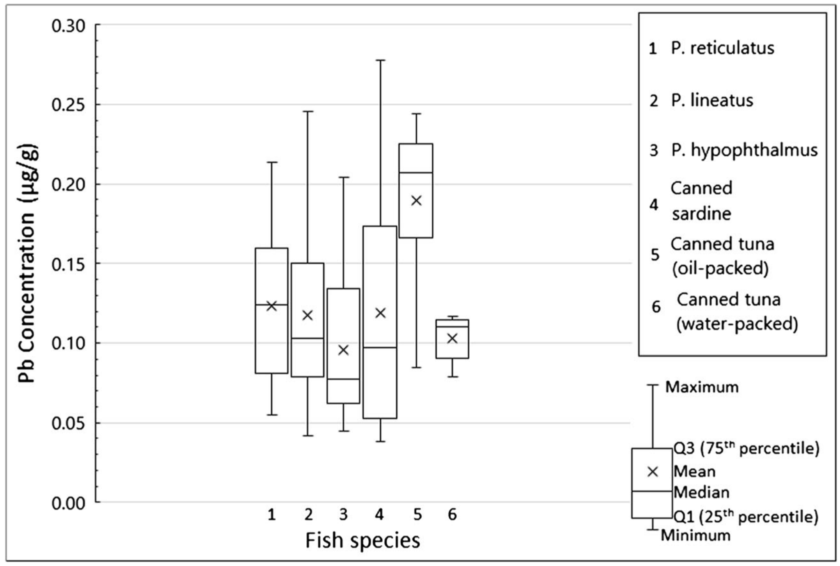


Fig. 1 Lead concentration in imported and marketed fish species

Pb is the most common metal identified in food, and in this case, in fish [19–21, 27]. According to Fig. 1, Pb levels among individual fish species were ordered as follows: canned tuna (oil-packed) > *P. reticulatus* > *P. lineatus* > canned sardines > canned tuna (water-packed) > *P. hypophthalmus*. For this metal, we found that most of the values remained below the standard limits (Table 1), although some samples had concentration values above the FAO/WHO limit (0.2 mg/kg fresh weight), except for the canned, water-packed tuna [29].

Cd may bioaccumulate in the human body and may induce kidney dysfunction, skeletal damage, and reproductive deficiencies [40, 41]. In this study, the Cd values in some samples were above all the reference standard limits (Table 1), but the mean values were below the 0.05 mg/kg limit, except for canned sardines (0.11 ± 0.09). The Cd concentration profile among species was as follows: canned sardines > *P. hypophthalmus* > *P. lineatus* > *P. reticulatus* > canned tuna (water-packed) > canned tuna (oil-packed), although canned sardines had a wide range of values, as did *P. hypophthalmus* and *P. lineatus* (Fig. 2).

The Zn concentration profile among species was as follows: canned tuna (oil-packed) > canned tuna (water-packed) > canned sardines > *P. lineatus* > *P. reticulatus* > *P. hypophthalmus*. Canned species concentrations showed a wide margin of difference compared to the other species (fresh and frozen), doubling, and even tripling, the Zn concentration values (Fig. 3). Although there is no maximum limit of Zn defined for fish or food in general, there are clear recommendations about its daily intake (DI), since the excessive consumption of zinc may cause some negative effects, such as dizziness, vomiting, anemia, and lethargy, which are the first symptoms of poisoning [42].

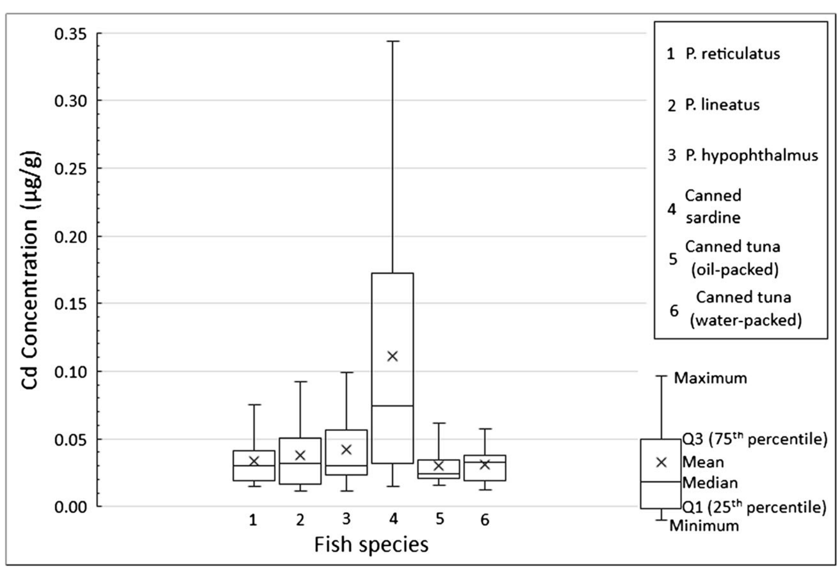
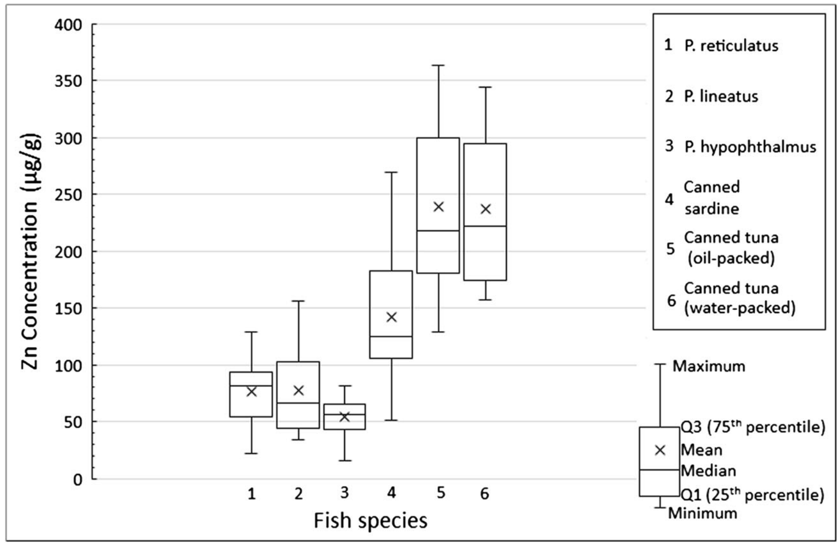


Fig. 2 Cadmium concentration in imported and marketed fish species

Fig. 3 Zinc concentration in imported and marketed fish species



The daily intake (DI) recommended for Zn by the Agency for Toxic Substances and Disease Registry (ATSDR) of the USA is 8 mg/day on average for women and 11 mg/day on average for men [42]. Despite the high average concentration values obtained in this study for canned species, the EDI values obtained for Zn are below the ATSDR recommendation (Table 2).

Comparing other studies worldwide, we observed that the values for Pb in our research were lower than the Pb values reported in other investigations [24, 27, 34, 38], except for in canned, oil-packed tuna, which had higher values in our study [36, 37] (Table 1).

For Cd concentrations, we found lower values in *Prochilodus* sp. than the values previously reported in other global studies [24]. We presume that the studied specimens were cultivated under controlled contamination conditions for exportation purposes. Regarding the mean concentration values obtained in this study for basa fillet, we assume that it is a cultured species [26] that is being imported and consumed in this coastal city (Barranquilla), and it is also within the

Table 1 Concentrations (mg/kg) of heavy metals in imported fish species reported in other studies worldwide

Fish species	Pb mg/kg	Cd	Zn	Country/reference
P. reticulatus Venezuelan “Bocachico”	7.15–10.29	0.85–3.4	–	Venezuela [24]
	0.12	0.03	76.1	This study
P. lineatus	8.06	–	52.68	Argentina [34]
Argentinian “Bocachico”	0.10	–	–	Argentina [35]
	0.12	0.04	77.1	This study
P. hypophthalmus	0.09	0.01	1.12	India [26]
Basa fillet	0.94	0.48		
	0.10	0.04	53.8	This study
Canned tuna	0.72	0.01–0.45	–	Ghana [27]
(water-packed)	0.10	0.03	236.6	This study
Canned tuna	0.04	0.02	–	Persian Gulf area [36]
(oil-packed)	0.1	0.08	–	Turkey [37]
	0.19	0.03	238.9	This study
Canned sardines	0.93–1.09	0.19–0.23	23.88–33.51	Brazil [38]
	0.09	0.19	–	Turkey [37]
	0.09	0.07	–	Kentucky, USA [39]
	0.12	0.11	141.6	This study
Permissible limit	0.2	0.05	–	FAO/WHO [29]
	0.3	0.05–0.1	–	EU [32]
	0.3	0.1	–	MHSP* [33]

*MHSP** Colombian Ministry of Health and Social Protection

Table 2 EDI (μg/kg/day), CR_{lim} (g/day), and MPI for heavy metals in different fish species for different groups

Species	EDI						CR_{lim}						MPI
	CHD			RP			CHD			RP			
	Pb	Cd	Zn	Pb	Cd	Zn	Pb	Cd	Zn	Pb	Cd	Zn	
P. reticulatus	0.19	0.05	120.5	0.05	0.03	60.3	1138.2	1029.4	138.0	2276.4	2058.8	276.0	0.71
P. lineatus	0.15	0.05	96.6	0.04	0.02	48.3	1196.6	921.1	136.3	2393.2	1842.1	272.5	0.72
P. hypophthalmus	0.13	0.06	74.5	0.03	0.03	37.2	1458.3	833.3	195.5	2916.7	1666.7	391.1	0.63
Canned tuna (water-packed)	0.12	0.04	275.2	0.03	0.02	137.6	1359.2	1129.0	44.4	2718.4	2258.1	88.8	0.92
Canned tuna (oil- packed)	0.22	0.03	277.9	0.05	0.02	139.0	740.7	1166.7	43.9	1481.5	2333.3	87.9	1.09
Canned sardines	0.16	0.15	187.9	0.04	0.08	93.9	1176.5	315.3	74.1	2352.9	630.6	148.3	1.20

CHD children, *RP* rest of the population

RfD: Pb, 4 μg/kg/day [30]; Cd, 1 μg/kg/day [43, 44]; Zn, 300 μg/kg/day [42, 43]

allowed standards for Cd content in fish. For canned species in this study, the average values for Cd in tuna were similar (both water-packed and oil-packed). They were within the permitted limits and had similar data to other investigations [27, 36]. However, we observed that there were reported values higher than the international limits [37].

For most cases in this and other similar studies, canned sardines were reported to contain higher values than were allowed for human consumption of Cd [37–39], suggesting a possible correlation between the type of fish and the canning process. Regarding Zn, we found few data for these species. This might be because it is a micronutrient and its intake is not internationally regulated. However, the values reported in other investigations are much lower than those reported in this study [26, 34, 37, 38]. Notably, all of the canned species reported quite high values compared to fresh and frozen species. We think there is a correlation between the canning process and the presence of elevated amounts of Zn and Cd in those samples. Other authors have reported that aspects such as the storage time, the type of varnish used, the thickness and porosity of the can, and the relative humidity in the air may influence the increasing metal content migration to food [45].

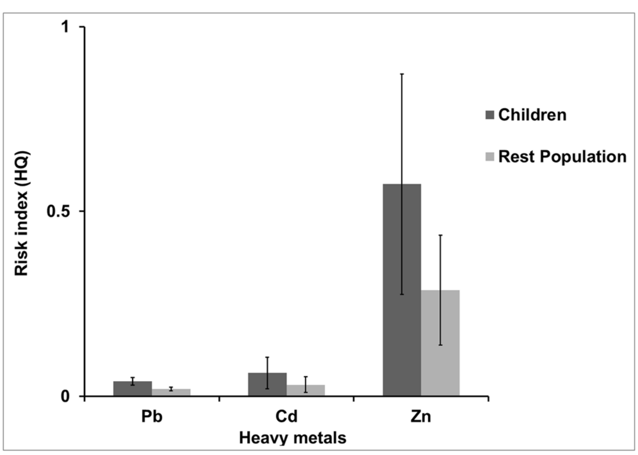


Fig. 4 Hazard quotients for total fish species for different groups. (CHD children, RP rest of the population)

Health Risk Assessment

The EDI results for this study were 0.12–0.22 for Pb, 0.03–0.15 for Cd, and 74.5–277.9 for Zn (Table 2). The highest values were for Zn, followed by Pb, and, finally, Cd. The children group (CHD) presented higher values than the rest of the population group (RP) at 4.1 times higher for Pb, 1.9 times higher for Cd and 2 times higher (double) for Zn. In addition, the EDI results for Zn in water-packed and oil-packed tuna were very close to the RfD for canned fish (300 μg/kg/day), and the reports for the maximum allowable fish consumption rate (CRlim) are also close to the average daily fish consumption for the CHD group at 40.7 g/day. These findings indicate that even though the different population groups had values below the permissible limits of the RfD established by the US EPA, the JECFA, and the ATSDR, the CHD group is at a higher potential health risk than the RP group, especially from the consumption of canned fish with heavy metals such as Cd and Pb, because these nonessential nutrients can bioaccumulate in the kidneys, liver, and muscles [46]. Additionally, the prolonged consumption of small amounts of heavy metals leads to the interruption of some physiological and biochemical processes in the body [47].

Hazard quotients (HQs) are used to assess the potential risk to human health with exposure to one or more heavy metals [43]. When HQ > 1 for any metal in food, it means that the consumer population faces a potential health risk. The higher HQ value was for Zn, and the lower value was for Pb in all of the evaluated samples for both adults (RP) and children (CHD) in this study (Fig. 4). The Zn values in the CHD group were approximately double those in the RP group. The HQs for the different metals were less than 1; however, it must be taken into account that the value for Zn was very close to the limit (HQ > 1) in the CHD group for canned, water-packed tuna (0.92), and oil-packed tuna (0.93). The values of the RP group were approximately 50% of the reference values, indicating that the consumption of fish represents a potential health risk from Zn, even if it is an essential element. The MPI presented higher results for the canned fish than for the fresh fish, indicating a greater metal accumulation in the samples with this packing for consumers (canned) (Table 2). The accumulation order based on MPI was sardines > tuna (oil-packed) > tuna (water-packed) > *P. lineatus* > *P. reticulatus* > *P. hypophthalmus*. These results are particularly concerning because canned fish meat is widely consumed due to its easy preparation, low price and nutritional value. High consumption can have significant adverse effects on human health due to the bioaccumulation of heavy metals [48]. This is why the consumption of fresh fish with lower MPI, such as *P. hypophthalmus*, for instance, is recommended.

Conclusions

The reported mean values for Pb, Cd, and Zn were below the suggested limits of the FAO, the EU, and the Colombian Ministry of Health and Social Protection (Table 1) for all the studied species. However, some samples had concentrations above the standard limits. When compared to the results of global studies, lower values were found in this research for Pb and Cd, but that was not the case for Zn.

There was no clear relationship between the trophic level and heavy metal bioaccumulation in this study. The probable cause of bioaccumulation is the contamination of the water bodies from which the fish come. On the other hand, canned fish (sardines and tuna) presented values that may suggest contamination problems with the canning process (pH, oxygen concentration in the headspace, quality of the inside lacquer coating of cans, storage place, storage time, etc.). The data showed that canned sardines had high values of Cd (0.111 ± 0.099), Pb (0.119 ± 0.073), and Zn (141.62 ± 60.84). Meanwhile, canned, oil-packed tuna had higher Pb values than the same water-packed species. This may suggest that the canning process in oil specifically may have an effect on the Pb content, which eventually is absorbed by the meat of the fish. Both types of canned tuna presented the same zinc levels (236.61 ± 67.41 in oil-packed and 238.93 ± 76.43 in water-packed), which was almost double or triple the values of this metal in the other four imported species. These results suggest a review of the canning processes and how they may contribute to heavy metal contamination in the different brands of tuna and sardines that are commercialized throughout the world.

The HQ for Zn was very close to exceeding the limit (HQ > 1) for water-packed (0.92) and oil-packed tuna (0.93) for the CHD group, while for the other two metals, the HQ was lower than 1. With these results, it can be affirmed that there is no health risk for the population in the short term since they did not exceed their RfDs. However, the HQ value for Zn indicates that the CHD group is at higher risk than the RP group in the long term because of the daily intake and the bioaccumulation of these metals over time, and because Pb and Cd were also detected, not only Zn, especially in canned fish.

MPI data also presented higher values for canned fish than for fresh or frozen fish, indicating, once again, that there might be a relationship between heavy metal accumulation and canned products. We suggest periodic monitoring of metals in these products because the coastal population of Barranquilla consumes them as an economical, protein-rich alternative food source that is easy to prepare.

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Compliance with Ethical Standards

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

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