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Evaluation of Mercury, Lead, Arsenic, and Cadmium in Some Species of Fish in the Atrato River Delta, Gulf of Urabá, Colombian Caribbean

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Abstract The concentrations of mercury, lead, cadmium, and arsenic were evaluated in 96 samples, 12 by each one of the following eight fish species: snook (*Centropomus undecimalis*), crevalle jack (*Caranx hippos*), Serra Spanish mackerel (*Scomberomorus brasiliensis*), southern red snapper (*Lutjanus purpureus*), blue runner (*Caranx crysos*), Atlantic tarpon (*Megalops atlanticus*), ladyfish (*Elops saurus*), and Atlantic goliath grouper (*Epinephelus itajara*), which were collected during 1 year in the Atrato River Delta in the Gulf of Urabá, Colombian Caribbean. Three fish were caught from each of the following sites the community usually uses to catch them (known as fishing grounds): Bahía Candelaria, Bahía Marirrío, Bocas del Roto, and Bocas del Atrato. The quantification of metals was performed by microwave-induced plasma-optical emission spectrometry. The Pb concentration fluctuated from 0.672 to 3.110 mg kg^{-1}, surpassing the maximum permissible limit (MPL = 0.3 mg kg^{-1}) for human consumption for all species. The Hg concentration ranged between < Limit of detection and 6.303 mg kg^{-1}, and in the crevalle jack and Atlantic tarpon, concentrations exceeded the MPL (0.5 mg kg^{-1}). The levels of Cd and As were not significant in the studied species and did not exceed the MPL (0.05 mg kg^{-1}).

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1 Introduction

Global fish production for human consumption increased steadily over the past five decades, parallel with the supply of edible fish, which increased at an average annual rate of 3.2% (between 1961 and 2013). Consequently, the average availability and apparent per capita fish consumption worldwide increased from 9.9 kg in 1960 to 14.4 kg in 1990 and to 19.7 kg in 2013, with projections for an even greater increase in subsequent years (FAO and Fisheries and Aquaculture Department 2016).

With the increase of fish consumption, exposure to the heavy metals that are present in the environment from the natural and anthropogenic factors has increased. The presence of these metals and organic compounds in water resources affects aquatic ecosystems and, therefore, human health (Leyva Cambar et al. 2010; Olivero Verbel and Johnson Restrepo 2002). When heavy metals are released into water resources, depending on the environmental conditions of the water (pH, redox potential and complexing agents), they accumulate in aquatic sediments and enter the tissues and organs of fish (Chandra Sekhar et al. 2004; Zhang et al. 2015), being the fish products, being a pollution vehicle for humans.

Among the most toxic metals are mercury (Hg), lead (Pb), cadmium (Cd), and arsenic (As); these metals can be found naturally in low concentrations in the environment. However, their levels have increased due to population growth and industrialization, thereby affecting the food chain and causing bioaccumulation and biomagnification in organisms that are unable to purge them (Jayaprakash et al. 2015; Lozada Zarate et al. 2007; Malik et al. 2010).

Hg is one of the most toxic heavy metals in the environment. The Hg affects the nervous system (Doadrio Villarejo 2004; Liu et al. 2008a; Muñoz Vallejo et al. 2012) and can also cross the blood-brain barrier and the placenta, which makes it neurotoxic and teratogenic (Clémens et al. 2011; Doadrio Villarejo 2004).

Pb is one of the most ubiquitous metals and is especially found in food and in the air. It interacts with essential metals such as calcium (Ca), iron (Fe), zinc (Zn), and copper (Cu), (Rubio et al. 2004; Valdivia Infantas 2006); its toxic effects are related to the central and peripheral nervous system and the gastrointestinal tract (Nava Ruíz and Méndez Armenta 2011; Rubio et al. 2004); and it is highly dangerous for the neonate because it also crosses the blood-brain barrier and the placenta (Castro González and Méndez Armenta 2008; Rubio et al. 2004).

For its part, Cd is easily absorbed into tissues and is deposited in the liver and, to a lesser extent, in the kidney (Liu et al. 2008b).

Finally, As is a metalloid classified by the International Agency for Research on Cancer (IARC) as a Group 1 carcinogen in humans (World Health Organization 2015). The natural exposure occurs with the consumption of contaminated water, fishery products, and seafood (Klaassen 2013).

Fish from the upper level of the food chain, such as those analyzed in this study, are more vulnerable in accumulating greater amounts of metals, which are absorbed following the ingestion of particulate matter suspended in water, the consumption of other metal-bearing fish or the exchange of dissolved metal ions through lipophilic membranes. This accumulation depends on both the absorption and elimination rates, and therefore, these metals are concentrated in different organs according to the metal-organ affinities (Squadrone et al. 2013). These piscivorous fish, because they are at the top of the food chain, become bioindicators as they can reflect the concentrations of metals in their surrounding environment.

Because fishery products are intended for human consumption, maximum limits of heavy metal residues in these products have been established. At present, national and international standards on maximum permissible limits (MPLs) exist for fishery products. In Colombia, the Ministry of Health and Social Protection established MPLs for Hg, Pb, and Cd of 0.5, 0.3, and 0.05 mg kg^{-1} of fresh weight, respectively, in Resolution 122 of 2012 (Ministerio de Salud y Protección Social de Colombia 2012). The European Community (Comisión de las Comunidades Europeas 2006; Comisión Europea 2011, 2014) and the World Health Organization (WHO)/Food and Agriculture Organization of the United Nations (FAO) (FAO/WHO 1972, 1989) have established similar maximum levels for Hg, Pb, and Cd in fresh weight, and for As, the Agência Nacional De Vigilância Sanitária of Brazil (Ministério da Saúde 2013), allows a maximum limit of 1.0 mg kg^{-1} of fresh weight.

In this study, the concentrations of Hg, Pb, Cd, and As were evaluated by a sensitive analytical technique such as microwave-induced plasma-optical emission spectrometry (MIP OES) (Herrero Fernández et al. 2014) in eight species of commercial importance, captured in the Atrato River Delta, Gulf of Urabá in the Colombian Caribbean.

2 Materials and Methods

2.1 Sampling

In total, 12 specimens of each of the eight most commercialized species in the area were caught, namely common snook (*Centropomus undecimalis*), crevalle jack (*Caranx hippos*), serra Spanish mackerel (*Scomberomorus brasiliensis*), southern red snapper (*Lutjanus purpureus*), blue runner (*Caranx crysos*), Atlantic tarpon (*Megalops atlanticus*), ladyfish (*Elops saurus*), and Atlantic goliath grouper (*Epinephelus itajara*), corresponding to three fish caught near or at the following four sites that the community usually use for fishing (known as fisheries): Candelaria Bay, Marirrío Bay, Bocas del Roto, and Bocas del Atrato. Ninety-six samples were obtained during 1 year (see Fig. 1). The sites that are located in Atrato River Delta, Gulf of Urabá in the Colombian Caribbean, on the border with Panama, can be affected by possible pollution sources (e.g., population density, urbanization, treatment plants), production activities (e.g., agricultural or mining production), transportation activities, and tourism activities. The Gulf is a semi-enclosed body of water with an area of 4.291 km^2, of which 650 km^2 corresponds to an estuarine area bordered by mangrove forests (Ortiz et al. 2003). The climate regime of the area is governed by the Intertropical Convergence Zone (ZCIT). It presents a dry period from the end of December until April and a wet period that begins in May. The daily average temperature fluctuates between 26 and 28 °C (Ortiz et al. 2003; Phillipe Chevillot et al. 1993) and that there are no seasons.

The number of samples was determined based on the considerations of the ethics committee for animal experimentation at the University of Antioquia, which required that the number of fish collected be minimized due to reduced populations of these organisms in recent decades. These species were captured under authorizations required by current regulations: permission framework was issued by the National Authority of Environmental Licenses (ANLA) in Resolution 0524 of May 27, 2014, and a fishing permit was issued by the National Authority for Aquaculture and Fisheries (AUNAP) in Resolution 00001827 on October 15, 2015. No need existed for prior consultation with the community according to the Ministry of Interior through office OFI14-000040724-DCP-2500 on October 31, 2014.

The captured organisms were adults with a minimum catch size that exceeded the average size at maturity, see Table 1. For the catch, experimental fishing was performed with gillnets of 50 m in length and 6–7 in. of eye mesh (bass-fishing net), thereby ensuring randomness of catch and protection of small species. The captured fish were then sacrificed in a bath with a 0.04% solution of eugenol and transported in a cooler with ice to the Laboratory of Marine Sciences, University of Antioquia, Turbo.

In the laboratory, the following characteristics were evaluated to determine the freshness of the fish: brightness, color, odor, and skin firmness. These samples were stored in resealable bags made of linear low-density polyethylene, and they were immediately frozen and stored in a freezer at − 20 °C. The samples were then transported by air in a cooler with freezer packs, thereby maintaining the cold chain, to the Diagnostic and Pollution Control Laboratory, where they were stored in a freezer at − 20 °C for later analysis.

2.2 Processing and Determination of Metals

For the quantification of heavy metals, the methodology according to AOAC 977.15 (as amended) was carried out for the determination of Hg (AOAC INTERNATIONAL 1978), and the methodology according to AOAC 999.11 (as amended) was carried out for the determination of Pb, Cd, and As (AOAC INTERNATIONAL 1999). Analyses were performed in triplicate, and the analytical results are expressed in mg kg^{-1} wet weight.

The acids were measured using a dispenser (Brand, 25 ml), Milli-Q-quality (Millipore, Bedford, MA, USA) water was used, and a certified glass was used for the digestions after being washed four times with a solution of 5% HNO_3 (Merck, Darmstadt, Germany) and then rinsed four times with deionized water. Homogenization was performed using a Hobart CC-34 industrial food processor (Hobart Troy, OH). A Shimadzu AUW120D balance (Shimadzu, Kyoto, Japan) was used for obtaining weights, which were accurate to less than 0.1 g. The digestions were performed on a Centricol hot plate with a water bath (Centricol, Medellin, Colombia). An Agilent 4100 spectrometer was used for sample measurement by MIP OES (Agilent, Santa Clara, CA), equipped with a multimode sample input system (MSIS) for Hg and As. For other metals, a OneNeb inert nebulizer with a double-pass chamber and an Agilent 4107 nitrogen generator were used, following the parameters described in Table 2. To verify the accuracy and precision, a certified reference material (CRM), SRM 1946 (Lake Superior Fish Tissue), which was

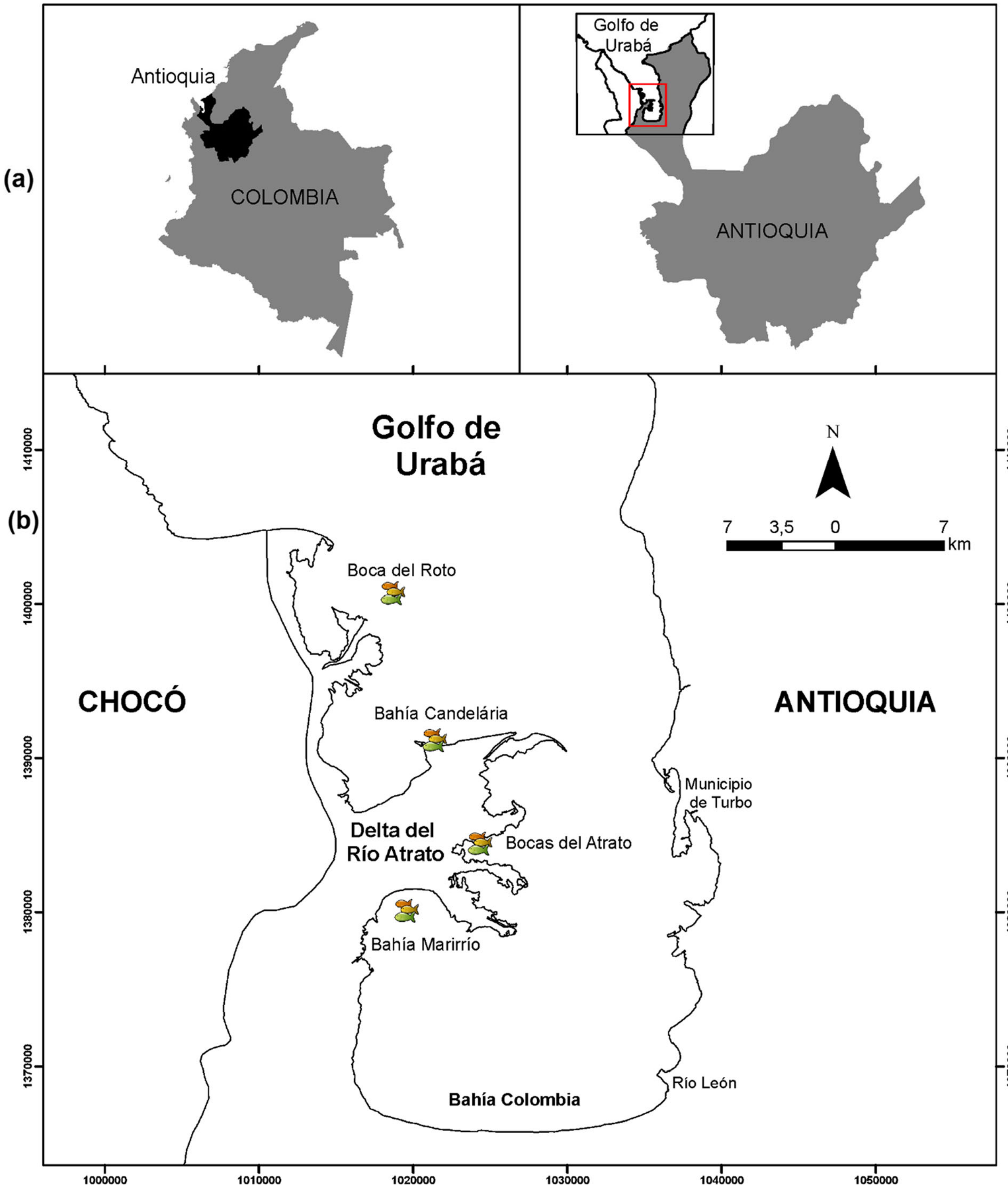

Fig. 1 **a** Location of the Gulf of Urabá in Colombia and the Caribbean region. **b** Location of the fishing grounds around which the catch was made in the Gulf of Urabá: Bocas del Roto, Candelaria Bay, Bocas del Atrato, and Marirrío Bay

purchased from the National Institute of Standards and Technology (NIST) in Maryland, USA, was used. All the details of the validation can be reviewed in the previous article (Gallego Ríos et al. 2017).

Table 1 Biometry of the fish studied

Species	Length (cm)	Height (cm)	Weight (kg)
Atlantic tarpon	100.250 ± 48.9232	33.958 ± 21.112	12.436 ± 15.466
Crevalle jack	88.283 ± 5.6756	24.792 ± 1.437	7.198 ± 0.823
Atlantic goliath	48.708 ± 9.7876	15.483 ± 3.874	1.570 ± 0.834
Southern red snapper	46.742 ± 7.8332	14.604 ± 3.096	1.418 ± 0.835
Common snook	43.150 ± 12.5997	10.492 ± 3.340	0.954 ± 0.539
Serra	50.667 ± 7.16473	9.025 ± 1.330	0.679 ± 0.348
Ladyfish	51.308 ± 4.96578	7.358 ± 1.123	0.581 ± 0.163
Blue runner	33.283 ± 3872	9.325 ± 1.880	0.342 ± 0.132

Values are expressed as mean ± SD

SD standard deviation

2.2.1 Hg

For the digestion, 1 g of the previously homogenized sample, contained in an Erlenmeyer flask in which the digestion was to be performed, was weighed on an analytical balance. A total of 4 ml of HNO_3, 2 ml of H_2SO_4, and 1 ml of HCl were added to the samples. The blank was made with the same amounts of acids that were added to the samples. The samples were placed in a water bath at 80 °C, where they remained for 2 h. The samples were allowed to cool for 1 h to prevent the release of vapors. After digestion, the sample was transferred to a 50-ml graduated volumetric flask, with deionized water added to make up the remainder of the volume. For the reference material (SRM 1946), half of these amounts were used for the sample, the acids and the final volume.

2.2.2 Pb, Cd, and As

For the digestion, 5 g of the previously homogenized samples, contained in Erlenmeyer flasks in which the digestions were to be performed, was weighed on an analytical balance. Ten milliliters of HCl and 5 ml of HNO_3 were added to the samples. The blank was made with the same amounts of acids that were added to the samples. Samples were heated on a hot plate at 140 °C for 2 h. After digestion, the samples were transferred to a 50-ml graduated volumetric flask, with deionized water being added to return the solution to volume. For the CRM, the amounts used for the sample, the acids and the final volume were halved.

2.3 Statistical Analysis

Statistical analysis was performed using Statgraphics Centurion XV (StatPoint Inc., USA). The valuation of the normality of the continuous variables was performed via the Shapiro-Wilk test. Nonparametric statistics was applied to those variables that were not normally distributed. A descriptive analysis was applied for heavy metal content using quantitative response measures of central tendency and of dispersion (mean, median, range, and standard deviation). An analysis of variance (ANOVA) was used to evaluate the existence of significant differences between the concentrations of heavy metals by species and site; if this analysis yielded a statistically significant difference, a post-ANOVA honestly significant difference (HSD) Tukey test was used for those variables that were normally distributed. In the case of those not normally distributed, a Kruskal-Wallis analysis was performed, followed by a comparative analysis of two groups via the Mann-Whitney *U* test for those variables with a statistically significant difference. For all statistical analyses, the criterion of significance was set at $p < 0.05$.

3 Results

The concentrations of metals (Hg, Pb, As, and Cd) in the eight species, as well as differences in the concentrations by collection sites, are presented for each metal in Tables 3, 4, and 5.

Table 2 Emission wavelength, detection limits, and summary of the certified and experimental values of the reference material SRM 1946

Metal	Wavelength (nm)	Detection limit (mg kg^{-1})	Standard reference material SRM 1946		
			Certified value (mg kg^{-1})	Found value (mg kg^{-1})	Recovery (%)
Hg	523.652	0.010	0.433 ± 0.009	0.430 ± 0.012	99.33
Pb	405.781	0.074	0.7[a]	0.679 ± 0.041	96.95
As	234.984	0.023	0.277 ± 0.010	0.242 ± 0.018	87.29
Cd[b]	228.802	0.011	–	–	98.6

[a] Information values are typically provided with no uncertainty because of the lack of sufficient information to adequately assess the uncertainty associated with a value

[b] Values corresponding to the accuracy data of the method

3.1 Hg

The greatest concentrations of Hg were found in crevalle jack, followed by Atlantic tarpon (see Table 3), with both exceeding the MPL of 0.5 mg kg^{-1}. Other species have concentrations below the maximum allowable limit. The Hg concentrations showed the following order from greatest to least: Crevalle jack > Atlantic tarpon > Serra Spanish mackerel > Blue runner > Ladyfish > Common snook > Southern red snapper > Atlantic goliath grouper (Fig. 2).

3.2 Pb

All species exceeded the MPL of 0.3 mg kg^{-1}. The Pb concentrations were found in a range of 1.181–2.744 mg kg^{-1}. The Ladyfish was the species with the highest average content, followed in descending order by the Blue runner > Atlantic tarpon > Southern red snapper > Atlantic goliath grouper > Serra Spanish mackerel > Common snook > Crevalle jack (Fig. 2), with the contents of the Serra Spanish mackerel and the crevalle jack being significantly lower compared to the other species (Table 4).

3.3 As

In the crevalle jack, Atlantic goliath grouper and Southern red snapper, As was not detected (see Table 5). The ladyfish was the species with the highest content of As, which did not exceed the MPL. The other species reported insignificant amounts (Fig. 2) but showed statistically significant differences among them because of the absence of As in most species ($p < 0.05$). Consequently, none of the species exceeded the MPL when taking into account the average levels of this metal.

Table 3 Differences between the concentrations of Hg by species and catch site

Species	Concentration (mg kg^{-1})	Median (IQR)	*p*-site
Crevalle jack(1)	1.230	3.4815	0.248†
Atlantic tarpon(2)	0.931	1.142	0.469*
Serra(3)	0.273 [1]	0.2885	0.650*
Blue runner(4)	0.149 [1 2 3]	0.0485	0.734*
Ladyfish(5)	0.133 [1 2 3]	0.098	0.537*
Common snook(6)	0.086 [1 2 3 4 5]	0.074	0.469*
Southern red snapper(7)	0.052 [1 2 3 4 5]	0.1	0.770†
Atlantic goliath(8)	0.054 [1 2 3 4 5]	0.044	0.055†

Values are expressed in mg kg^{-1} wet basis

IQR interquartile range

$p < 0.05$, *ANOVA with Tukey or † Kruskal-Wallis posttest

The concentration column indicates differences among the species to which the number corresponds

Table 4 Differences among the Pb concentrations by species and catch site

Species	Concentration (mg kg^{-1})	Median (IQR)	*p*-site
Ladyfish$^{(1)}$	2.744	1.326	0.975†
Blue runner$^{(2)}$	2.028	2.064	0.319†
Atlantic tarpon$^{(3)}$	2.015	2.067	0.859†
Southern red snapper$^{(4)}$	1.781	2.1	0.933†
Atlantic goliath$^{(5)}$	1.322	1.945	0.972†
Serra$^{(6)}$	1.304	1.798	0.740†
Common snook$^{(7)}$	1.472 ^{1}	1.528	0.450*
Crevalle jack$^{(8)}$	1.181 ^{1}	0.462	0.108*

Values are expressed in mg kg^{-1} wet basis

IQR interquartile range

$p < 0.05$, *ANOVA with Tukey or † Kruskal-Wallis posttest

The concentration column indicates differences among the species to which the number corresponds

3.4 Cd

For this study, of all the samples analyzed, only 6% had some level of concentration, but the concentration was insignificant and well below the MPL of 0.05 mg kg^{-1}. Cd concentrations for ladyfish and Serra Spanish mackerel were even below the limit of quantification 0.04.

4 Discussion

In the present study, no significant concentrations of As and Cd were found in any of the fish. The Pb contents exceeded the MPL for all species of fish. Hg was also found in all fish; however, the crevalle jack and Atlantic tarpon were the only species in which Hg levels exceeded the MPL. These results could be related to the anthropogenic activity in the study area, in this there are urban centers and tourist areas lacking wastewater treatment systems and landfills near water sources. There is a port for navigation, in addition to industrial fishing. There is also legal and illegal mining of gold and coal in the tributary river basins and industrial activity of cardboard, plastics, and agricultural (banana) (Ruiz Muñoz et al. 2012).

In the area where the research was carried out, there are no studies on the concentrations of Hg, Cd, Pb, and As in fish; however, there are several monitoring

Table 5 Differences among the concentrations of As per species and catch site

Species	Concentration (mg kg^{-1})	Median (IQR)	*p*-site
Ladyfish$^{(1)}$	0.133	0.154	0.397†
Atlantic tarpon$^{(2)}$	0.001	0.240	0.247†
Serra$^{(3)}$	< LOD	0.221	0.668†
Common snook$^{(4)}$	< LOD	0.218	NA
Blue runner$^{(5)}$	< LOD	0.1125	NA
Crevalle jack$^{(6)}$	< LOD $^{1\ 2\ 3\ 4\ 5}$		NA
Atlantic goliath$^{(7)}$	< LOD $^{1\ 2\ 3\ 4\ 5}$		NA
Southern red snapper$^{(8)}$	< LOD $^{1\ 2\ 3\ 4\ 5}$		NA

Values are expressed in mg kg^{-1} wet basis

IQR interquartile range

$p < 0.05$, *ANOVA with Tukey or † Kruskal-Wallis posttest

The concentration column indicates differences among the species to which the number corresponds

LOD limit of detection

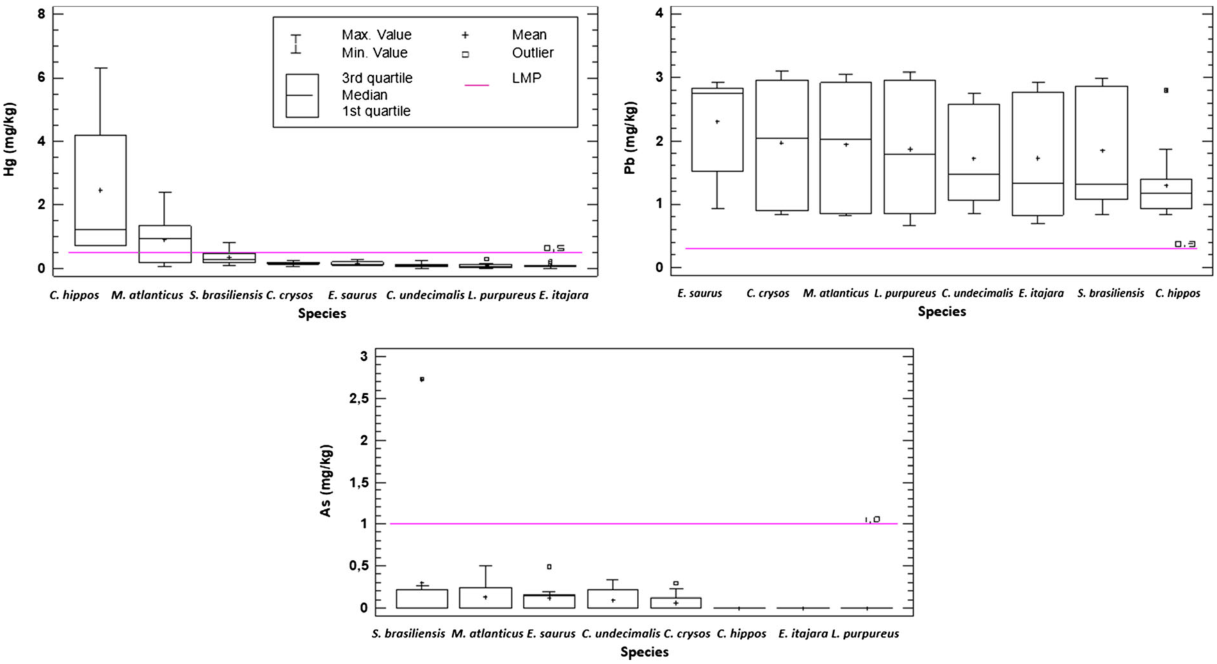

Fig. 2 Comparison of the concentrations of Hg, Pb, and As (mg kg^{-1}) among the species caught in the Gulf of Urabá

stations of sea water of INVEMAR (Institute of Marine and Coastal Research) that monitor the concentrations of heavy metals. The last monitoring of the year 2008 reported an average value of Hg in water of 9.29 μg/l (INVEMAR and Ministerio de Ambiente Vivienda y Desarrollo Territorial 2008), concentration above the quality criteria established by the legislation of reference, for the existence and permanence of Hg in Water corresponding to 0.1 μg/l (INVEMAR and Ministerio de Ambiente y Desarrollo Sostenible 2014). Also reported, Pb levels of 11.5 μg/l (INVEMAR and Ministerio de Ambiente Vivienda y Desarrollo Territorial 2008) are high but not above the norm.

4.1 Hg

The highest Hg concentrations were found in crevalle jack (1.230 mg kg^{-1}), followed by Atlantic tarpon (0.931 mg kg^{-1}). These results are consistent with reports for crevalle jack (*Caranx hippos*) in Suriname, which had concentrations of 1.17 ± 0.27 μg g^{-1} (Mol et al. 2001). However, other studies of this species in different parts of the world (Emmanuel and Samuel 2009; Voegborlo and Adimado 2010), show lower concentrations for Hg without exceeding the MPL of 0.5 mg kg^{-1}, including the research in Cartagena (Colombia) where average concentrations of 0.09 ± 0.03 μg g^{-1} were obtained (Olivero Verbel et al. 2009).

The high concentrations found for Hg in these two species may be attributed to them being larger, longer-lived fish, factors that can influence the bioaccumulation of this metal. Other characteristics can also affect bioaccumulation, including biological variability (physiology and diet), geological influences (sediments), chemical variability (water quality and biogeochemistry of Hg), physical variability (water temperature), the way in which methylmercury (MeHg) is processed or stored by certain species of fish, eating habits, swimming patterns, metal content in food, and other influences, such as time and atmospheric precipitation (El Moselhy et al. 2014; Licata et al. 2005; Voegborlo et al. 2007).

For this study, the concentrations of Hg in Serra Spanish mackerel were higher than the values reported by other researchers (Adamas and McMichael 2007; Mansilla-Rivera and Rodríguez-Sierra 2011; Mol et al. 2001; Thera and Rumbold 2014) as well as nine times greater than those reported by López Barrera in fish for sale in Bogota (Colombia), including *Scomberomorus* sp. (López Barrera and Barragán Gonzalez 2016). Stratified sampling was used for that study, which included wholesale and retail distribution centers such as markets, hypermarkets, supermarket chains, and convenience stores that sell local and imported products.

Concentrations of Hg in the blue runner (*Caranx crysos*) in this study were similar to those found in Florida (USA) (Thera and Rumbold 2014) and were even lower than those found in the Greater Accra (Ghana) (Voegborlo and Adimado 2010).

Concentrations for ladyfish of the same genus but of a different species, *Elops machnata,* found in Taiwan, were 38 times higher (Huang et al. 2008; Liao et al. 2016) than those in this study. Possibly this occurs because, although the species are in the same genus, the distribution ranges are different, with *Elops saurus* feeding on prey exposed to different environmental conditions, as *Elops saurus* dwells only in the Atlantic and *Elops machnata* only in the Indo-Pacific West. Notably, concentrations of Hg in *Elops saurus* analyzed in Cartagena (Colombia) were three times lower (Olivero Verbel et al. 2009) than those in this study.

For the common snook (*Centropomus undecimalis*), lower concentrations were found than in the study in Suriname (Mol et al. 2001), but previously mentioned research, carried out in Cartagena and Bogota (Colombia), reported concentrations similar to those of *Centropomus undecimalis* (0.09 ± 0.04 μg g^{-1}) (Olivero Verbel et al. 2009) and lower than those in *Centropomus* sp. (0.041 ± 0.0263 mg kg^{-1}) (López Barrera and Barragán Gonzalez 2016).

The concentrations for Southern red snapper (*Lutjanus)* of the same genus but different species in different regions of the world had similar concentrations (Mansilla-Rivera and Rodríguez-Sierra 2011; Mol et al. 2001; Thiyagarajan et al. 2012; Voegborlo and Adimado 2010), including those analyzed in Cartagena, which showed average concentrations of 0.08 ± 0.01 μg g^{-1} (Olivero Verbel et al. 2009).

The Hg concentrations in crevalle jack and Atlantic tarpon exceed the MPL of 0.5 mg kg^{-1} by up to five times and two times, respectively. Therefore, the consumption of these species could present a health risk, considering that the main Hg intake is eating contaminated fish. For this reason, controlling the intake of Hg by reducing edible portions is advisable, particularly for pregnant women, children, and the elderly, who are more sensitive to exposure to this metal. The region of Urabá is affected by mining of gold and other minerals and by water pollution resulting from municipal, residential, and agricultural effluents (Al Sayegh Petkovšek et al. 2012; Castro González and Méndez Armenta 2008; de Jesus et al. 2014; Doadrio Villarejo 2004; INVEMAR and Ministerio de Ambiente y Desarrollo Sostenible 2014; Liu et al. 2008a; Patterson 2002). However, this situation does not fully explain the content of Hg in the crevalle jack and Atlantic tarpon because they are migratory species, and the levels found in them were not necessarily obtained only from pollution in the Gulf. In contrast, levels reflect accumulations over their long lives and over different migratory routes.

4.2 Pb

The results of this study require special attention because the concentrations of Pb in all species tested exceeded the MPL of 0.3 mg kg^{-1} by up to eight times. The highest concentrations were found in ladyfish (2.744 mg kg^{-1}), without significant differences being noted among the other species, except for the Serra Spanish mackerel and the crevalle jack. The concentration of Pb in the blue runner was higher in this study than in other investigations from various regions of the world (El Moselhy et al. 2014; Khaled 2009; Renata J. Medeiros et al. 2012; Renata Jurema Medeiros et al. 2014). Additionally, in studies carried out on the Southern red snapper, we found concentrations up to ten times higher than those in the Red Sea (El Moselhy et al. 2014), Pearl River (China) (Leung et al. 2014), Tamil Nadu (India) (Thiyagarajan et al. 2012), and Greece (Psoma et al. 2014) as well as in common snook captured in Vitória Bay (Joyeux et al. 2004) and Sao Paolo (Morgano et al. 2011) in Brazil and in Bogotá (Colombia) (López Barrera and Barragán Gonzalez 2016), with concentrations in our study being 7, 11, and 59 times higher, respectively. Similar results were found for both Atlantic goliath grouper (El Moselhy et al. 2014; Leung et al. 2014; Mok et al. 2012; Siavash Saei-Dehkordi and Fallah 2011; Thiyagarajan et al. 2012) and Serra Spanish mackerel (Khaled 2009; López Barrera and Barragán Gonzalez 2016; Siavash Saei-Dehkordi and Fallah 2011; Zhu et al. 2015) compared to other places in the world.

Pb contamination in the Gulf can be influenced by the establishment of large urban centers that host populations with deficient water treatment systems for domestic and industrial wastewater, the presence of landfills close to water sources, different industries (cardboard, plastic, and agricultural) that utilize Pb or its derivatives in their processes, and the influence of legal and illegal gold and coal mining in the upper middle and lower basins of the Atrato river. However, as with Hg, this situation does not fully explain the content of Pb in the studied species because they are migratory, and their levels reflect Pb that has been accumulated during their long lives along different routes.

4.3 As

No species exceeded the MPL with its average level of As. The As concentrations were below the limit of quantification for Crevalle jack, Atlantic goliath grouper, and Southern red snapper, and even though the Ladyfish was the species with the highest content (0.133 mg kg^{-1}), its concentration was also below the MPL.

The species with the highest average concentrations was the Serra Spanish mackerel, with levels similar to other studies (Mansilla-Rivera and Rodríguez-Sierra 2011; Zhu et al. 2015). However, concentrations of the study carried out at wholesale and retail sale sites in Bogota for *Scomberomorus* sp. and *Centropomus* sp. reported levels six and two times below (López Barrera and Barragán Gonzalez 2016) than those in this study. This result might be because, in the latter, researchers were only able to identify the genus of the specimens studied, which means that feeding patterns and accumulation in the said specimen might be very different from those in the present study. The concentrations in the blue runner relate to the results of other investigations for *Caranx crysos*, which were higher (Renata J. Medeiros et al. 2012; Renata Jurema Medeiros et al. 2014) than those in this study.

So far, the amounts present for this metal do not pose a health risk. However, the fish from this study showed traces of this metal, and a future increase in the rate of release of As from sulfide minerals derived from excavation activities can occur as a result of the continuation of current mining practices (Wang and Mulligan 2006).

4.4 Cd

Although some possible sources of Cd contamination occur in the area, such as the use of certain phosphate fertilizers, manure, and blue bags in banana plantations (Faroon et al. 2012; Ruiz Muñoz et al. 2012; Zuluaga Rodríguez et al. 2015), these sources seem not to influence the concentrations of Cd in fish. The amounts of this metal found in the fish were well below the maximum limits, and detection was not even possible in some of the samples.

5 Conclusions

The highest concentrations of Hg were found in crevalle jack and Atlantic tarpon, both with concentrations that exceeded the MPL, possibly due to ingestion of the metal for prolonged periods and to high intramuscular fat content, which tended to accumulate greater amounts of Hg. In addition, these species are characterized by their large size, longevity, migratory behavior, and wide distribution in the Gulf of Urabá, which would imply a greater exposure to possible sources and greater bioaccumulation and biomagnification of this metal.

All species exceeded the MPL for Pb due to the many factors that can affect the contamination of water, whether from a natural source (the crust); from landfill disposal near bodies of water; or from direct dumping of domestic, agricultural, and industrial wastewaters as well as from the influence of legal and illegal gold and coal mining, to which these species are exposed along their migratory routes.

Concentrations of Cd and As in most species were well below the MPL and, in some cases, were below the limit of quantitation (LOQ).

Because of the harmful effects of heavy metals in aquatic ecosystems and the consequences of these metals on the health of both animals and humans, controls must be carried out. Thus, the oversight of concentrations of heavy metals by competent authorities is recommended in both potentially contaminated areas and edible species. This oversight should include risk assessments to estimate the potential impact of these pollutants on human health, and in this way, oversight authorities as well as fishermen, farmers, and consumers can make appropriate decisions based on scientific evidence.

This information is intended to contribute to the current management process that is provided by the competent authorities, in which the involvement of all stakeholders in the fisheries sector will lead to the proposal of practices that will contribute to pollution reduction in the Gulf of Urabá.

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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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