Human and Ecological Risk Assessment: An International Journal

ISSN: 1080-7039 (Print) 1549-7860 (Online) Journal homepage: https://www.tandfonline.com/loi/bher20

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HUMAN AND ECOLOGICAL RISK ASSESSMENT
https://doi.org/10.1080/10807039.2018.1529552

Metals and arsenic in marine fish commercialized in the NE Brazil: Risk to human health

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ABSTRACT

Arsenic, cadmium, lead, and mercury in fish is the result of long-term biomagnification in the food chain and is of public concern, due to the toxicity they engender. The objective of this research was to determine the concentrations of arsenic, cadmium, lead, and mercury in 13 species of marine fish broadly commercialized in Aracaju, SE, Brazil and to evaluate the risks of fish consumption associated with these trace elements, using the Target Hazard Quotient (THQ). As, Cd, and Pb levels were measured with inductively coupled plasma mass spectrometry (ICP-MS), and mercury was analyzed via cold vapor atomic absorption spectrometry. The results indicate a large variability in concentrations for arsenic (0.07–2.03 mg kg^{-1}) and mercury (0.01–1.44 mg kg^{-1}), associated with the animal dietary category. Cadmium (0.04–0.19 mg kg^{-1}) and lead (<0.01–0.45 mg kg^{-1}), on the other hand showed a mild variability. None of the evaluated specimens had As, Cd, and Pb THQ values higher than 1. The THQ values for mercury were higher but indicated no consumption risk, except for amberjack, and snook fish. Overall THQ indicates lower risk of consumption in fish that are at the base of the food chain, than in those that are top predators.

ARTICLE HISTORY
Received 3 September 2018
Revised manuscript accepted 25 September 2018

KEYWORDS
toxicity; metals; target hazard quotient; marketed fish; maximum tolerance limits; health risk

Introduction

Fish meat is rich in essential amino acids, vitamins, micro- and macroelements, and ômega-3 long-chain polyunsaturated fatty acids such as docosahexaenoic acid and eicosapentaenoic acid, and is also low in cholesterol (Guil-Guerrero *et al.* 2011). These characteristics confer on fish meat the advantage of qualifying as one of the more healthful foods (Storelli 2008; Afonso *et al.* 2013; Farrugia *et al.* 2015); its consumption is beneficial for neurological growth and development, and it reduces the risk of stroke disease and the incidence of cardiovascular diseases, while also enhancing the human immune response (He 2009; Pohlenz and Gatlin 2014). In contrast to the dietary

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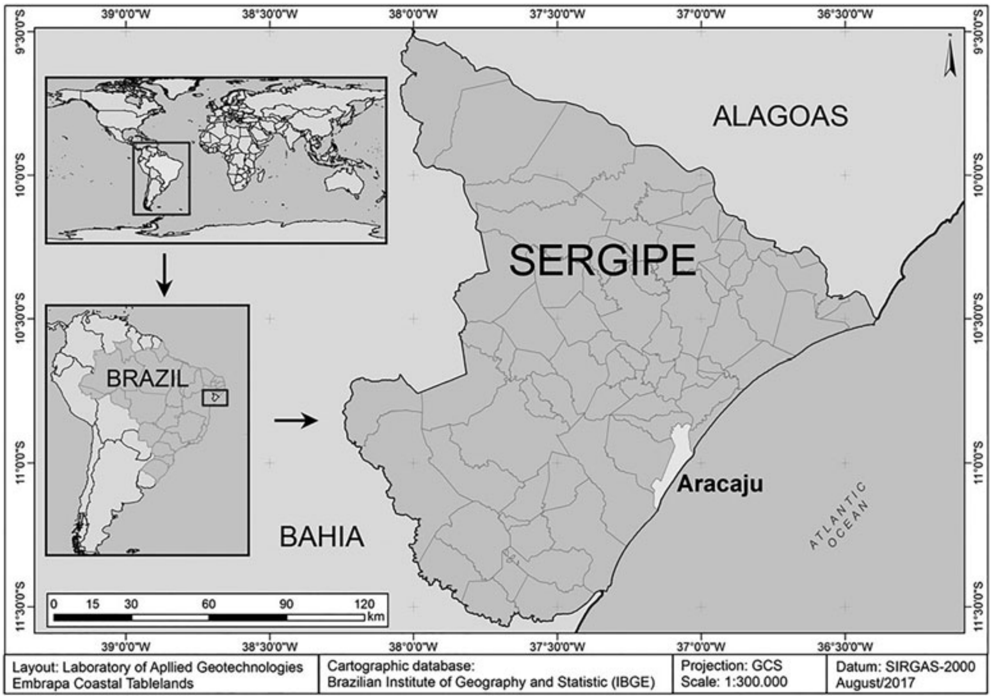

Figure 1. Study area. Location of the Municipality of Aracajú in the State of Sergipe, Brazil.

advantages of fish, however, the potential risk of exposure to chemical contaminants (and their derivatives) contained in fish must also be considered, when assessing the influence of this food on human health (Bosch *et al.* 2016). In fact, there has been an increasing reluctance to eat fish because of reports stating that its consumption can result in significant exposure to a variety of chemicals. Despite the uncertain risk/benefit tradeoff, demand for fish products is increasing worldwide while wild-caught landings remain stable (FAO 2016).

Trace metals stand out among chemical pollutants because of their ability to accumulate in the tissues of aquatic organisms and the consequent possibility of contaminating humans through the food chain (Castro-González and Méndez-Armenta 2008; Mathews and Fisher 2008, 2009, Guo *et al.* 2016; Schneider *et al.* 2018). The trace elements arsenic, cadmium, lead, and mercury present in fish meat at parts-per-million (ppm) levels are considered toxic, and can cause deleterious effects on human health (Rainbow 2007; Berk *et al.* 2014).

Fish containing concentrations higher than the background level of these trace elements have been measured by environmental protection agencies using risk assessment tools. The Target Hazard Quotient (THQ) estimation methodology for human health risk assessment was initially proposed by the United States Environmental Protection Agency in the 1980s (USEPA 1989). The THQ is based on the calculation of metal concentrations in food, intake rates, and the reference oral dose, and has been used in studies evaluating the potential risk of exposure to contaminants in vegetables (Yañez *et al.* 2018), vegetable oils (Zhu *et al.* 2011), oysters (Zhao *et al.* 2016), and fish (Yi *et al.* 2011; Adel *et al.* 2016; Avigliano *et al.* 2016; Gu *et al.* 2017).

Because of the potential and/or real danger of consumption of fish with significant levels of toxic metals, many countries monitor the levels of these trace elements in fish locally consumed, to protect human health. In Brazil, the National Health Surveillance Agency (ANVISA) is responsible for the control of food contaminants through the Maximum Tolerance Limits (MTL) for inorganic contaminants, published in Resolution # 42, dated August 29, 2013 (ANVISA 2013). The values of MTL for metals in fish and their products are: 1.00 mg kg^{-1} arsenic; 0.30 mg kg^{-1} lead; from 0.05 to 0.30 mg kg^{-1} cadmium, depending on the species; and 0.50 mg kg^{-1} mercury for non-carnivorous fish and 1.00 mg kg^{-1} mercury for carnivores.

However, surveillance of contaminant levels above the MTL prescribed by legislation is not sufficient to characterize the risk to human health, because fish intake rate and frequency of exposure to the contaminant are not considered in the MTL. Furthermore, consumers generally do not have enough information to make decisions about what kind of fish to eat, considering that they do not know how the levels of contaminants vary among fish.

Considering the risk of contamination in humans, through fish consumption, concentrations of arsenic, cadmium, lead, and mercury in 13 species of marine fish commercialized in Aracaju were determined and quantified and compared with the MTL prescribed by the Brazilian Legislation. The fish consumption risks associated with these trace elements was evaluated, using the Target Hazard Quotient tool.

Material and methods

Sample collection and preservation

The fish species to be evaluated were selected as a function of information from the Fisheries and Aquaculture Statistical Bulletin (Brasil 2011) and from a fish market study in Aracaju, Sergipe, Brazil (Anonymous 2004), as well as on their availability and purchase frequency among the general population.

Samples of 13 species of marine fish were randomly purchased at general-purpose markets, fish markets, street markets and/or supermarkets in Aracaju, Sergipe, northeastern Brazil (Figure 1), from March to April 2016, totaling 39 samples, with 3 samples per species. The classification, habitat, and dietary categories of these fish are shown in Table 1. The fish samples were transported refrigerated in thermal boxes to Embrapa Coastal Tablelands Laboratory in Aracaju, SE. Approximately 100–150 g of the lateral–dorsal muscle of each fish were separated and lyophilized for 48 h, until they reached constant weights. Afterwards, they were stored in a freezer at –15 °C. Samples were ground in a processor and sieved through 250 μm nylon mesh to obtain homogeneous samples. Between each milling the processor was washed with 10% v/v nitric acid solution and then with Milli-Q water (18 $\mu\Omega$) to avoid cross-contamination between samples.

Reagents and standard solutions

All solutions were prepared with analytical-grade reagents, and the Milli-Q water (18 $\mu\Omega$) was obtained from a Millipore® Simplicity®UV (Molsheim, France) purifier.

Table 1. Classification, habitat, and dietary category of the studied fish species.

Common name	Family	Scientific name	Habitat	Dietary category
Amberjack	Carangidae	*Seriola* spp.	Demersal	Carnivorous
Tuna fish	Scombridae	*Thunnus* spp.	Pelagic	Piscivorous
Catfish	Ariidae	*Bagre* spp.	Demersal	Benthic invertebrate feeder
Dolphinfish	Coryphaenidae	*Coryphaena hippurus*	Pelagic	Fish and invertebrate feeder
Acoupa weakfish	Sciaenidae	*Cynoscion acoupa*	Demersal	Fish and benthic invertebrate feeder
Weakfish	Sciaenidae	*Cynoscion* spp.	Demersal	Fish and benthic invertebrate feeder
Sardine	Clupeidade	*Sardinella brasiliensis*	Pelagic	Planktivorous
Mullet	Mugilidae	*Mugil curema*	Pelagic	Planktivorous
Snapper fish	Lutjanidae	*Lutjanus* spp.	Demersal	Omnivorous
Snook	Centropomidae	*Centropomus undecimalis*	Neritic	Piscivorous
Cobia	Rachycentridae	*Rachycentron canadum*	Benthic and reef-associated	Fish and benthic invertebrate feeder
Whitemouth croaker	Sciaenidae	*Micropogonias furnieri*	Neritic	Benthic invertebrate feeder
Grouper	Serranidae	*Mycteroperca interstitialis*	Benthic and reef-associated	Piscivorous

All materials used in the preparation and analysis of the samples were immersed in 10% v/v HNO_3 acid bath for 24 h and rinsed with Milli-Q (<18 $\mu\Omega$) water prior to use. Standard stock solutions of As, Cd, Pb (100 mg L^{-1}), and Hg (1000 mg L^{-1}) SpecSol® (Jacareí, Brazil) were used in the preparation of the calibration standards.

Sample digestion

The metals contained in about 0.4–0.5 g (dry weight) of lyophilized triplicate samples of muscle were extracted in a closed microwave system Anton Paar, model Multiwave 3000, at the maximum power of 1400 W, in the four steps represented in Table 2. The extraction was carried out with 10 mL nitric acid (7 M) and 2 mL 30% v/v hydrogen peroxide (H_2O_2), according to methodology adapted from Jarić *et al.* (2011). After cooling to room temperature, the digested samples were brought to the volume of 100 mL with Milli-Q water (<18 $\mu\Omega$) and stored at 4 °C until analysis.

For Hg analyses, triplicate 0.5 g of the sample were digested with 15 mL 1:1 solution of (H_2SO_4+HNO_3) in a kinetic reactor (cold finger), heated for 2 h in a 60 °C sand bath. After cooling, 3 mL H_2O_2, 52 mL deionized water, and 15 mL 5% potassium permanganate ($KMnO_4$) were added. The excess oxidant was then neutralized with 12% hydroxylamine hydrochloride (Hight and Cheng 2005), and 5 mL of the sample were placed in the reaction flask with 1 mL stannous chloride $SnCl_2$ (20%) (Hatch and Ott 1968).

Detection methods and quality control

An inductively coupled plasma mass spectrometer (ICP-MS, Thermo, Germany) was used for the quantitative determination of As, Cd, and Pb. The instrumental parameters of ICP-MS were: radiofrequency applied power of 1.3 kW, plasma gas flow rate of

Table 2. Microwave digestion system operational parameters.

Step	Temperature (°C)	Ramp (min)	Hold (min)	Fan level
1	100	13.0	7.0	Slow
2	150	7.0	12.0	Slow
3	180	6.0	10.0	Slow
4	50		30.0	Fast

13 L min^{-1}, auxiliary gas flow rate of 0.7 L min^{-1}, nebulizer gas flow rate of 0.87 L min^{-1}, peak jump scan mode, residence time of 10 ms, and number of readings per replicate was equal to 3. Reagent blanks were processed in the same manner as the samples and were read every ten sample batteries. Total mercury reading was carried out in a Lumex RA 915+ cold vapor atomic absorption spectrophotometer (CVAAS, with Zeeman correction, Russia).

The validation of the analytical method was done with reference material DORM-3 (fish protein certified reference material for trace metals—NRCC) and the recovery values of the analytes warranted the quality of the data.

Risk assessment

Mean concentrations of arsenic, cadmium, lead, and mercury were used for the estimation of the *THQ*. The formula used in the *THQ* calculation was proposed by the USEPA (1989) as Eq. (1):

$$THQ = \left(EF \times ED \times FIR \times C/RFD \times BW \times ET\right) \times 10^{-3} \tag{1}$$

where *EF* is exposure frequency (365 days year^{-1}); *ED* is exposure duration (70 years), equivalent to the average human-life estimate; *FIR* is food intake rate (fish =36 g person^{-1} day^{-1}, as suggested by USEPA (1989); *C* is the metal concentration in the fish (μg g^{-1}); *RFD* is the oral reference dose (As $=0.3 \times 10^{-3}$ μg g^{-1} day^{-1}, Cd $=1 \times 10^{-3}$ μg g^{-1} day^{-1}, Pb $=4 \times 10^{-3}$ μg g^{-1} day^{-1}, Hg $=0.5 \times 10^{-3}$ μg g^{-1} day^{-1}) (Storelli 2008; USEPA 2010); *BW* is the mean adult body weight (70 kg); *ET* is the non-cancerous exposure time (365 days year^{-1}×ED).

In the risk assessment, we assumed that the oral intake of the contaminant was equal to the dose absorbed by the human organism and that cooking the fish did not change the concentration or toxicity of the contaminant (USEPA 1989). If the calculated *THQ* is <1 there is no appreciable risk for a given pollutant; if the value of *THQ* is >1 there is an imminent risk to continuing to consume fish at that frequency (Storelli 2008).

Results and discussion

The precision and accuracy of the analytical method given by the recovery values of the analytes were within the range of 80–110%, considered acceptable for trace metal analysis (Table 3).

The concentrations of metals in the fish samples are given in mg kg^{-1} on a wet basis (Table 4) for comparison with values published on the same basis by the Brazilian Legislation.

Table 3. Concentrations of arsenic, cadmium, lead, and mercury (mg kg^{-1} dry basis) in the reference material DORM-3 ($n = 3$).

Element	Certified value (mg kg^{-1})	Measured value (mg kg^{-1})	Recovery (%)
As	6.88 ± 0.30	6.12 ± 0.61	88.95
Cd	0.29 ± 0.02	0.27 ± 0.03	93.10
Pb	0.39 ± 0.05	0.39 ± 0.08	100.00
Hg	0.38 ± 0.06	0.39 ± 0.01	102.63

Arsenic

The lowest and highest concentrations of total arsenic were 0.07 mg kg^{-1} in *Cynoscion* spp. and 2.03 mg kg^{-1} in *R. canadum*, respectively. The mean concentration in all fishes evaluated in this study was 0.69 ± 0.31 mg kg^{-1}, considerably lower than in fish of the Gulf of Mexico, averaging 4.23 ± 9.39 mg kg^{-1} in the reference area and 7.33 ± 14.25 mg kg^{-1} in the area impacted by pulp mill effluent (Lewis *et al.* 2002). Much higher concentrations were observed in the surroundings of an industrial complexes clustered along the coast in the central Adriatic Sea, where average of 41.17 ± 7.49 mg kg^{-1} for fish was observed (Perugini *et al.* 2014). Jureša and Blanuša (2003) also found high arsenic levels in fish along the Croatian coast of the Adriatic Sea: ranging from 0.56 to 23.30 mg kg^{-1}.

The concentrations range of As observed in this study was also lower than that found by Burger and Gochfeld (2005), which ranged from 0.23 to 3.30 mg kg^{-1} in marine fish, marketed in New Jersey, USA; 35% of those samples exceeded the maximum limit of 1.3 mg kg^{-1} prescribed by U.S. legislation (Fallah *et al.* 2011). In Brazil, a study conducted by the Food Technical Commission (CTA) in 2013 detected arsenic in samples of tuna fish, whitemouth croaker, and sardines marketed in São Paulo, SP, Brazil, with only 2.5% of the samples below the maximum tolerated values of 1.0 mg kg^{-1} set by Brazilian Legislation (CTA 2013). Medeiros *et al.* (2012) detected levels of total As between 0.002 and 11.800 mg kg^{-1} when analyzing fish commercialized in the main market of the city of Niterói, RJ, Brazil, whereas Silva *et al.* (2016) found between 0.48 and 1.19 mg kg^{-1} in fish commercialized in Salvador, BA, Brazil. In this study, guachanche barracuda (*Sphyraena guachancho*) and snapper fish (*Lutjanus synagris*) exceeded the maximum tolerable limit (MTL) of 1.0 mg kg^{-1} established by Brazilian Legislation. According to Burger and Gochfeld (2005), the comparison and interpretation of metal levels among commercial fish samples is difficult because their geographical origins are undetermined, and their diet may exhibit variations in both predation patterns and specificity. We believe that differences in the trace elements' content occurred because their accumulation in marine organisms is related not only to the presence of the pollutant, but also to a range of biological (species, age, growth stage) and environmental (temperature, geochemical anomalies, salinity) factors that influence incorporation processes (Burger *et al.* 2014). In American Samoa, the concentrations of total As in mullet (*Mugil* spp.) have ranged from 0.37 to 0.94 mg kg^{-1} (Peshut *et al.* 2008), while in the Black Sea concentrations ranged from 0.38 to 1.10 mg kg^{-1} (Makedonski *et al.* 2017). Those values are higher than observed in this study for mullet (*M. curema*), which ranged from 0.15 to 0.18 mg kg^{-1}.

The predatory tuna fish, snook, and cobia had mean levels of total arsenic above the maximum tolerable levels of 1.0 mg kg^{-1}, and two species—catfish and whitemouth

Table 4. Mean concentrations of metals (mg kg^{-1} w.w.) in 13 species of marine fish ($n = 3$) commercialized in Aracaju.

Species	Arsenic	Cadmium	Lead	Mercury
Amberjack *Seriola* spp.	0.56 ± 0.37 (0.17–1.00)	0.08 ± 0.02 (0.06–0.11)	0.04 ± 0.05 (0.03–0.10)	1.10 ± 0.01 (1.09–1.10)
Tuna fish *Thunnus* spp.	1.30 ± 0.34 (0.81–1.80)	0.08 ± 0.02 (0.06–0.14)	0.09 ± 0.06 (0.02–0.19)	0.60 ± 0.15 (0.47–0.76)
Catfish *Bagre* spp	0.62 ± 0.43 (0.11–1.25)	0.08 ± 0.03 (0.05–0.19)	0.03 ± 0.01 (0.02–0.05)	0.67 ± 0.02 (0.65–0.69)
Dolphinfish *Coryphaena hippurus*	0.58 ± 0.22 (0.32–0.76)	0.09 ± 0.02 (0.06–0.14)	0.23 ± 0.15 (0.11–0.45)	0.85 ± 0.04 (0.82–0.88)
Acoupa weakfish *Cynoscion acoupa*	0.69 ± 0.29 (0.46–1.02)	0.07 ± 0.03 (0.04–0.10)	<0.01	0.68 ± 0.01 (0.67–0.69)
Weakfish *Cynoscion* ssp.	0.22 ± 0.17 (0.07–0.44)	0.08 ± 0.02 (0.04–0.10)	0.03 ± 0.03 (0.02–0.10)	0.30 ± 0.05 (0.26–0.37)
Sardines *S.brasiliensis*	0.34 ± 0.17 (0.22–0.48)	0.08 ± 0.01 (0.07–0.10)	<0.01	0.06 ± 0.02 (0.03–0.09)
Mullet *Mugil curema*	0.16 ± 0.02 (0.15–0.18)	0.07 ± 0.04 (0.04–0.14)	0.13 ± 0.12 (0.04–0.21)	0.01 ± 0.00 (0.01–0.01)
Snapper fish *Lutjanus* spp	0.33 ± 0.10 (0.24–0.48)	0.07 ± 0.01 (0.06–0.10)	0.11 ± 0.08 (0.03–0.19)	0.62 ± 0.18 (0.46–0.72)
Snook *C. undecimalis*	1.07 ± 0.37 (0.81–1.33)	0.10 ± 0.04 (0.05–0.14)	<0.01	1.24 ± 0.16 (1.16–1.44)
Cobia *R. canadum*	1.46 ± 0.81 (0.88–2.03)	0.08 ± 0.00 (0.08)	<0.01	0.37 ± 0.03 (0.33–0.39)
Whitemouth croaker *M. furnieri*	0.95 ± 0.41 (0.57–1.39)	0.07 ± 0.02 (0.06–0.08)	<0.01	0.44 ± 0.02 (0.42–0.46)
Grouper *M. interstitialis*	0.58 ± 0.15 (0.42–0.70)	0.08 ± 0.01 (0.07–0.09)	0.08 ± 0.03 (0.05–0.11)	0.93 ± 0.22 (0.790–1.18)

Mean ± standard deviation; variation interval in parentheses.

croaker—presented some specimens with concentrations close to the MTL. The highest levels of As in this group of predators may be related to the carnivorous feeding habits (Table 1) that concentrate this element through the food chain. The high levels of As in tuna can be attributed to their piscivorous diet of pelagic plankton fish. Its prey feed on marine phytoplankton, which has the ability to accumulate inorganic arsenic and transform it into organic arsenic via methylation, which is then transferred to the tuna through the trophic chain (Eisler 1988; Li *et al.* 2003). Peshut *et al.* (2008) reported that total As analysis was not a good indicator for evaluating the toxicity of the element in humans, and emphasized the importance of quantifying the toxic inorganic fractions and nontoxic organic fractions. In speciation studies of fish tissues, Gao *et al.* (2018) and Ruelas-Inzunza *et al.* (2018) established that the nontoxic organic form arsenobetaine constituted around 90% of total arsenic in fish.

Cadmium

The lowest and the highest concentrations of cadmium measured in the fish were 0.04 mg kg^{-1} in the acoupa weakfish (*C. acoupa*) and weakfish (*Cynoscion* spp.), and 0.19 mg kg^{-1} in the catfish (*Bagre* spp.) and mullet (*M. curema*), respectively. The mean concentration of Cd in all fish in this study were 0.08 ± 0.02 mg kg^{-1}, lower than the mean values of 0.23 mg kg^{-1} Cd for the Gulf of Mexico (Lewis *et al.* 2002), but higher than the values for Black Sea fish on the Bulgarian coast (0.01 mg kg^{-1}) (Makedonski *et al.* 2017); South China Sea fish (0.02 mg kg^{-1}) (Gu *et al.* 2015); Atlantic Sea fish (0.02 mg kg^{-1}) (Chahid *et al.* 2014); Portuguese coast fishes (0.005 mg kg^{-1}) (Afonso *et al.* 2013); and Central Adriatic Sea fish (0.06 ± 0.04 mg kg^{-1}) (Perugini *et al.* 2014). The mean levels of Cd in tuna fish, sardines, and mullet did not exceed the MTL of 0.10 mg kg^{-1} specified for these species. The others—amberjack, catfish, dolphinfish, weakfish, snapper fish, snook, cobia, whitemouth croaker, and grouper fish—presented average levels above the MTL of 0.05 mg kg^{-1} prescribed by Brazilian Legislation.

Medeiros *et al.* (2012) analyzed 11 fish species commercialized in the main market of the city of Niterói, RJ, Brazil, and detected levels of Cd between 0.002 and 0.500 mg kg^{-1}. On the other hand, Morgano *et al.* (2011) found concentrations of Cd ranging from <0.010 to 0.287 mg kg^{-1} in weakfish (*Macrodon ancylodon*), mullet (*Mugil liza*), and sardine (*S. brasiliensis*) commercialized in São Paulo, SP, Brazil, similar to the mean concentration of Cd in tuna in this study (0.08 ± 0.02 mg kg^{-1}). These concentrations are higher than in measured in *Thunnus obesus* (0.043 ± 0.006 mg kg^{-1}) from South China Sea (Gu *et al.* 2017).

The mean Cd levels of 0.07 ± 0.04 mg kg^{-1} in mullet (*M. curema*) obtained in this study were higher than those found in another species of mullet (*M. cephalus*) which averaged 0.012 ± 0.002 mg kg^{-1}, from South China Sea by Gu *et al.* (2015). Zaza *et al.* (2015) reported values of Cd from 0.03 to 0.11 mg kg^{-1} in fish collected from Eastern Central Atlantic fishing areas and Afonso *et al.* (2013) observed concentrations of 0.02 mg kg^{-1} in various Portugal coast fishes. Joyeux *et al.* (2004) analyzed fish from the bay of Vitória, ES, Brazil, and found values between 0.02 and 0.04 mg kg^{-1} in *Mugil* spp.—lower than the 0.04–0.14 mg kg^{-1} obtained for this species in this study.

Similarly, cadmium content in this fish from the basin of São Francisco do Conde, BA, Brazil varied from 0.01 to 0.08 mg kg^{-1} (Santos *et al.* 2013).

Lead

The lowest and highest concentrations of lead observed in this study were below detection limits in acoupa weakfish, sardines, snook, cobia, and whitemouth croaker, and were 0.45 mg kg^{-1} in dolphinfish. The mean concentration of lead for all the fish in this study was 0.09 ± 0.07 mg kg^{-1}, higher than the 0.07 ± 0.02 mg kg^{-1} reported by Lewis *et al.* (2002) for fish from the Gulf of Mexico, of 0.05 mg kg^{-1} by Chahid *et al.* (2014) for Atlantic Sea fish, of 0.04 ± 0.02 mg kg^{-1} by Perugini *et al.* (2014) for Central Adriatic Sea fish, and by Guérin *et al.* (2011), who found 0.011 ± 0.009 mg kg^{-1} in fish collected in a French market. Our values were also lower than the 0.36 mg kg^{-1} observed by Gu *et al.* (2015) in South China Sea fish. The dolphinfish (*C. hippurus*) was the only species evaluated that showed levels of lead above the MTL set by Brazilian legislation (0.30 mg kg^{-1}). The differences in Pb concentrations among species can be attributed to the type of food consumed and the differences in fish eating habits (Serrão *et al.* 2014). Pb concentrations similar to those found in this study were observed for 11 species of fish traded in Niterói, RJ, Brazil, which presented levels between 0.01 and 0.50 mg kg^{-1} (Medeiros *et al.* 2012), between 0.03 and 0.48 mg kg^{-1} in fish commercialized in São Paulo, SP, Brazil (Morgano *et al.* 2011), and from 0.09 to 0.40 mg kg^{-1} in samples of canned fish collected in markets from Turkey (Tuzen and Soylak 2007).

Guérin *et al.* (2011) reported average levels of lead from 0.024 to 0.047 mg kg^{-1} for sardine, higher than those obtained in this study, whereas Elnabris *et al.* (2013) found higher levels (0.55 ± 0.48 mg kg^{-1}) in whitemouth croaker commercialized in Palestine. On the other hand, Zaza *et al.* (2015) reported values from 0.05 to 0.17 mg kg^{-1} in fish collected from Eastern Central Atlantic fishing area. Burger and Gochfeld (2005) found levels above 0.2 mg kg^{-1} in tuna marketed in New Jersey, USA—higher than those obtained in this study.

Compared with other studies, the Pb levels of 0.13 ± 0.12 mg kg^{-1} in the *M. curema* obtained in this study were lower than the 0.25 ± 0.05 mg kg^{-1} reported for *Mugil brasilienses* from Baia de Todos os Santos, BA, Brazil (Santos *et al.* 2013) and the 0.17 ± 0.09 mg kg^{-1} in mullet marketed in Palestine (Elnabris *et al.* 2013). A study by Silva *et al.* (2016) found values ranging from 0.019 to 0.022 mg kg^{-1} in sea bream (*Archosargus rhomboidalis*) and snapper (*L. synagris*) marketed in Salvador, BA, Brazil, while in the Mediterranean, Storelli *et al.* (2013) observed Pb concentrations ranging from 0.08 to 0.12 mg kg^{-1}, considered safe for consumption.

Mercury

The lowest (0.005 mg kg^{-1}) and highest (1.440 mg kg^{-1}) Hg concentrations were found in *M. curema* and *C. undecimalis*, respectively. The average concentration of mercury in planktivorous fish was 0.03 ± 0.01 mg kg^{-1}, whereas in the predator group, it was 0.71 ± 0.07 mg kg^{-1}. Hg levels in planktivorous species did not exceed the MTL of 0.5 mg kg^{-1} determined by Brazilian legislation (ANVISA 2013). The highest mercury

Table 5. *THQ* values for fish consumption, for inorganic arsenic, cadmium, lead, and mercury, in Aracaju, Brazil.

Species	Inorganic arsenic[a]	Cadmium	Lead	Mercury	*TTHQ*[b]
Amberjack	0.10	0.04	0.00	1.13	1.27
Tuna fish	0.22	0.04	0.01	0.62	0.89
Catfish	0.10	0.04	0.00	0.69	0.83
Dolphinfish	0.10	0.05	0.03	0.87	1.05
Acoupa weakfish	0.12	0.03	0.00	0.70	0.85
Weakfish	0.03	0.04	0.00	0.31	0.38
Sardines	0.05	0.04	0.00	0.06	0.15
Mullet	0.03	0.03	0.02	0.00	0.08
Snapper fish	0.05	0.03	0.01	0.64	0.73
Snook	0.19	0.05	0.00	1.28	1.52
Cobia	0.26	0.04	0.00	0.38	0.68
Whitemouth croaker	0.15	0.03	0.00	0.45	0.63
Grouper	0.10	0.04	0.01	0.81	0.96

Values above 1.00 indicate risk.
[a]The inorganic content constitutes 10% of the total As in fish, according to Liao and Ling (2003) and Lin *et al.* (2005).
[b]$TTHQ = THQAs + THQCd + THQPb + THQHg$.

level of 1.24 ± 0.16 mg kg^{-1} was observed in *C. undecimalis*, a carnivorous species from the top of the chain that, in the adult phase, feeds preferentially on fish (De Sousa Pereira *et al.* 2015), contributing to an extensive biomagnification of Hg (Kehrig *et al.* 2017). The *C. undecimalis* and *S. lalandi* were the species that presented Hg levels above the MTL of 1.0 mg kg^{-1} specific for carnivorous fish (ANVISA 2013; Esposito *et al.* 2018). According to Storelli (2008) and Ahmad *et al.* (2015), carnivorous species that feed on the bottom, such as snook and amberjack, present a great tendency to incorporate more mercury than those that feed close to the surface, such as tuna fish. This is due to the higher availability of Hg at the water-sediment interface, caused by the continuous production of methylmercury in this system (Bratkič *et al.* 2017). Nonetheless, after Farrugia *et al.* (2015) a reduction in mercury toxicity is attained when fish present higher concentrations of selenium.

Compared to other studies, the average level of Hg in *S. brasiliensis* (0.06 ± 0.02 mg kg^{-1}) in this study was similar to the mean of 0.053 ± 0.003 mg kg^{-1} in *Sardinella aurita* marketed in São Paulo, SP, Brazil (Augelli *et al.* 2007). Our results were higher than the 0.019 ± 0.011 mg kg^{-1} in sardine also commercialized in São Paulo, SP, Brazil (Morgano *et al.* 2011) and lower than the 0.130 ± 0.045 mg kg^{-1} in sardine from Cabo Frio, RJ, Brazil (Silva *et al.* 2011), the 0.08 ± 0.05 mg kg^{-1} in *S. brasiliensis* from Ilha Grande Bay, RJ, Brazil (Seixas *et al.* 2015). The mean concentration of 0.006 ± 0.001 mg kg^{-1}in *M. curema* in this study was lower than the 0.012 ± 0.007 mg kg^{-1} in *M. liza* marketed in São Paulo, SP, Brazil (Morgano *et al.* 2011) and the 0.015 ± 0.001 mg kg^{-1} in mullet from Guanabara Bay, RJ, Brazil (Kehrig *et al.* 2002). Zaza *et al.* (2015) reported average Hg values of 0.082 ± 0.026 mg kg^{-1} in *Tilapia heudelotii* collected from an Eastern Central Atlantic fishing area.

The mean level of Hg of 0.60 ± 0.10 mg kg^{-1} in *Thunnus* spp. observed in this study is similar to the 0.66 ± 0.10 mg kg^{-1} reported by Burger and Gochfeld (2005) in similar fish species marketed in New Jersey, USA. Our values were higher than the 0.35 mg kg^{-1} in tuna fish destined for the Italian market (Galimberti *et al.* 2016); the 0.31 mg kg^{-1} in tuna fish marketed in Sesimbra, Portugal (Cabañero *et al.* 2005); and the 0.14 to 0.21 mg kg^{-1} in yellowfin tuna (*Thunnus albacares*) caught in two different geographic sites in the eastern Pacific Ocean (Ordiano-Flores *et al.* 2011). These results express the importance of the trophic position, the quality and composition of the preferred natural food intake, and longevity in the process of bioaccumulation and biomagnification of Hg, in the different species analyzed. The results obtained confirm that lower levels of mercury are generally observed in planktivorous fish than in carnivores (Kehrig *et al.* 2013; Ahmad *et al.* 2015; Galimberti *et al.* 2016).

Risk assessment

In any risk assessment of a population, realistic fish intake rates are required, in order to characterize the threat to human health (Watanabe *et al.* 2003). In this study, the fish intake or consumption rate was 36 g inhabitant^{-1} day^{-1} (as suggested by USEPA 1989), which corresponds to an apparent per capita consumption of 13.14 kg inhabitant^{-1} year^{-1}. This per capita consumption of fish is similar to that published by De Oliveira Sartori and Amancio (2012), of 12.8 kg inhabitant^{-1} year^{-1} for the population of the Northeast region of Brazil.

The evaluation of the levels of trace metals in locally consumed fish is the first step in evaluating the risk to the human population. In calculating the specific *THQ* for each element, associated with the consumption of fish, the average concentration of each metal in the fish muscle, which is the edible part, was considered. Table 5 presents the *THQ* values for the evaluation of the potential risks of arsenic, cadmium, lead, and mercury exposure, from the target species in the study.

Arsenic

The toxic effects of arsenic depend on the oxidation state, chemical species, exposure dose, solubility in biological media, and excretion rate. The chemical form is the main factor for determining the risks to human health, and inorganic arsenic is more toxic than the organic forms (ATSDR 2007; Santos *et al.* 2013). Arsenic forms vary greatly depending on the organism, environment, and geographic location (Maher *et al.* 2018). Because of this variability, the toxicity of arsenic to humans is not accurately assessed when risk analyses are limited to total arsenic, and it is necessary to determine the inorganic toxic and organic nontoxic fractions (ATSDR 2007). There is general consensus in the literature that about 85–90% of the arsenic in the edible parts of marine fish is organic arsenic (e.g., arsenobetaine) (USEPA 2003; Lin and Liao 2008). The THQ_{As} values of Table 5 were calculated based on the estimated toxic inorganic fraction, assuming that the inorganic arsenic constitutes 10% of the total arsenic in fish, similar to the calculations performed by Liao and Ling (2003) and Lin *et al.* (2005).

The arsenic THQ values (THQ_{As}) ranged from 0.03 to 0.26 in the fish commercialized in Aracaju (Table 5). None of the fish evaluated in this study had THQ_{As} values >1, and hence this does not constitute a risk for Aracaju consumers.

The high THQ_{As} values observed in cobia, tuna fish, and snook can be attributed to their higher trophic levels and piscivorous feeding habits, where the arsenic is transferred to the links succession of the trophic web (Gao *et al.* 2018). Sardine—the preferred prey of the tuna fish—presented higher arsenic content than the weakfish and mullet in our study, similar to the results reported by Morgano *et al.* (2011). Cobia had the highest THQ_{As} value of 0.26, which can be attributed to its benthic feeding habits, where these fishes are exposed to higher levels of sediment-associated contaminants than are pelagic fish (Storelli 2008; Perugini *et al.* 2014). When THQ is <1, there is no potential risk of harm to human health (USEPA 1989).

Cadmium

The health implications of cadmium exposure are worsened by the relative inability of humans to excrete cadmium. Acute Cd poisoning is characterized by fever, irritation of the eyes, nose and throat, cough, dyspnea, weakness, nausea, vomiting, abdominal cramps, and diarrhea, and may cause acute lung edema (Gu *et al.* 2017). Chronic exposure causes respiratory problems, dental cavities, yellowing of teeth, anorexia, fatigue, weight loss, pallor, anemia, proteinuria, and renal tubular damage (Godt *et al.* 2006; Dural *et al.* 2007). The cadmium THQ values (THQ_{Cd}) in the fish commercialized in Aracaju varied from 0.03 to 0.05 (Table 5), and the mean value of THQ_{Cd} was 0.04 ± 0.01 higher than the 0.02 ± 0.01 found by Gu *et al.* (2015). Storelli (2008) obtained THQ_{Cd} values between 0.01 and 0.04 for 18 marine fish species: ranges similar to those obtained in this study. The highest THQ_{Cd} of 0.05 was found in dolphinfish and snook, and all the THQ_{Cd} values for cadmium in this study were far <1, similar to those obtained by Gu *et al.* (2015) for four fish species from the South China Sea.

Lead

The adverse effects caused by the accumulation of lead in organisms are neurological damage and renal diseases, as well as cardiovascular and reproductive impairments (Alessio *et al.* 2007). The inorganic form is the most frequent, as well as the most toxic and most easily absorbed by organisms (Garza *et al.* 2006).

The values of the lead THQs (THQ_{Pb}) ranged from 0.00 to 0.03, and were similar to those measured by Storelli (2008) (between 0.002 and 0.180), and the mean value of 0.01 ± 0.01 was lower than that calculated by Gu *et al.* (2015) (0.02 ± 0.01) in the fish of the South China Sea. The highest value of THQ_{Pb}—0.03—was observed in the dolphinfish.

The THQs of lead and cadmium (Table 5) presented values <1 for all the target species in the study, and do not represent a risk to consumers of these fish. These results corroborate those obtained by Vieira *et al.* (2011) in fish commercialized in Porto, Portugal, who reported an absence of adverse effects on human health due to the levels of lead and cadmium found in the evaluated fish. Similar results were reported by Watanabe *et al.* (2003), who used other risk indicators and determined that cadmium and lead did not present any appreciable risk, although in their study, arsenic did show potential risk via fish consumption.

Mercury

The principal dietary source of neurotoxic mercury compounds is the ingestion of species of fish in which methylmercury has accumulated (González-Estecha *et al.* 2014).

Methylmercury from fish has been linked to neurological damage (Minamata disease) (Hong *et al.* 2012), adverse neurodevelopmental (Myers *et al.* 2009), cardiovascular (Grandjean *et al.* 2004), and immunological health effects (Gardner *et al.* 2010). The mercury THQ values (THQ_{Hg}) in this study ranged from 0.00 to 1.28, and the highest value was observed in *C. undecimalis*, followed by *Seriola* spp. (1.13), indicating potential risk to human health. The interval of variation in THQ_{Hg} observed in this study was similar to those reported by Storelli (2008) in European anchovy *Engraulis encrasicholus* and albacore *Thunnus alalunga* (from 0.08 to 1.87). On other hand, THQ_{Hg} was <1 in yellowfin tuna (*Thunnus albacares*) caught in the eastern Pacific Ocean (Ordiano-Flores *et al.* 2011) and was 0.21 in sharks from the Persian Gulf (Adel *et al.* 2016).

Two species of predatory fish, *Coryphaena hippurus* (0.87) and *Mycteroperca interstitialis* (0.81), presented THQ_{Hg} values close to 1, and the lowest values were observed in *M. curema* and *S. brasiliensis* (Table 5). These data corroborate Storelli (2008), who indicated a lower potential risk of consumption based on THQ in species at the base of the food chain than in predators at the top.

TTHQ

Some contaminants may have a synergistic effect when present in the same tissues and consumed together. Thus, Total Target Hazard Quotient (*TTHQ*) represents the simple sum of each specific *THQ* for each metal (Eq. (2)) as proposed by Chien *et al.* (2002):

$$TTHQ = THQ_{As} + THQ_{Cd} + THQ_{Pb} + THQ_{Hg} \quad (2)$$

The lowest and highest values of TTHQ were 0.08 in mullet and 1.52 in snook. The mean value of *TTHQ* was 0.77 ± 0.41 higher than the 0.64 calculated by Saha *et al.* (2016), the 0.35 found by Adel *et al.* (2016), and the 0.05 found by Gu *et al.* (2015), without considering the contribution of the risk associated with mercury. Storelli (2008), considering *THQ* relative to Hg, recorded a $TTHQ > 1$ in albacore *Thunnus alalunga* (2.08), rosefish *Helicolenus dactylopterus* (1.54), and thornback ray *Raja clavata* (1.06). The fish in this study that presented a potential risk to the human health based on $TTHQ > 1$ were amberjack, dolphinfish, and snook (Table 5).

Conclusions

The levels of lead in the muscle of the target species presented acceptable toxicological levels for human consumption, except for dolphinfish, which presented samples that exceeded the MTL established by Brazilian Legislation. Regarding the cadmium levels, only tuna fish, sardines, and mullet did not exceed the MTL; however, based on *THQ*, none of the evaluated fish presented a risk to the consumer.

Tuna fish, snook, and cobia presented total arsenic levels above MTL, but none of the species evaluated had $THQ > 1$, calculated based on estimated inorganic arsenic, and did not represent a potential risk to human health. Speciation for arsenic compounds is necessary to determine the organic (nontoxic) and inorganic (toxic) forms in the fish and to enable more accurate risk assessments. In addition, it is fundamental that Brazilian legislation establishes the maximum limits of inorganic arsenic levels in the different fish species.

Snook and amberjack presented total mercury contents above the MTL, and grouper presented values very close to this limit. The mean concentrations of total mercury were lower in the fish at the base of the food chain and increased toward higher trophic levels. The need for speciation is relevant especially when, as in the specific cases of mercury and arsenic, the chemical forms demonstrate different impacts and behaviors such as toxicity, mobility and bioavailability.

The *THQ* estimate used in this study was sensitive enough to detect differences in potential human health risk, within the fish species and metals evaluated. *TTHQ* values >1, in descending order, accounted for: snook > amberjack > dolphinfish. These fish should be moderately consumed because of the potential human health risk caused by the evaluated metals. The reassessment of target species in future years is important to verify whether there is a change in the potential consumption risk for these species.

Knowledge about the potential risk of consumption of fish that may present arsenic, cadmium, lead, or mercury levels above the MTL is of major importance for health surveillance agencies, so that they can develop recommendations of safe levels of consumption of target species, in particular for children, pregnant women, and nursing mothers.

Funding

This study was supported by the Brazilian Council of Scientific and Technological Development [grant number 481925/2013-9]; the Foundation for the Support to the Research and Technological Innovation of the State of Sergipe; granted a Junior Research scholarship to SOS; the Foundation Carlos Chagas Filho for the Support to the Research of the State of Rio de Janeiro granted a Junior Research Scholarship to GCdP [grant number 216606]; the Brazilian Council of Scientific and Technological Development granted a research fellowship to JCW [grant number 306714/2013-2].

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