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Metal Levels in Fish Captured in Puerto Rico and Estimation of Risk from Fish Consumption

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Abstract Exposure to metals through fish consumption may represent a health risk, especially for high-fish-consumption populations such as fishing communities in the Jobos Bay and La Parguera areas in Puerto Rico. This study determined levels of As, Cd, Cu, Pb, Hg, Se, and Zn in muscle tissues of fish from the Jobos Bay and La Parguera (reference site) areas and estimated the health risk posed by fish ingestion to local fishermen and their children. Fish collected included *S. cavalla* ("sierra"; $n = 14$), *M. undulatus* ("roncón"; $n = 21$), *L. synagris* ("arrayado"; $n = 18$), and *L. analis* ("sama"; $n = 11$) in the Jobos Bay area and *S. regalis* (sierra; $n = 10$) and *L. synagris* (arrayado; $n = 8$) in La Parguera. Only As and Hg were detected at levels of human health concern. Average As and Hg levels (μg/g, wet wt) in the four species of Jobos Bay were 0.74 and 0.10 for roncón, 0.83 and 0.09 for sama, 1.00 and 0.26 for sierra, and 2.49 and 0.15 for arrayado, respectively. In La Parguera, average As and Hg levels (μg/g, wet wt) were 0.61 and 0.12 for sierra and 1.27 and 0.20 for arrayado, respectively. At both sites, the species with the highest As levels was arrayado, while for Hg, sierra obtained the highest concentrations. A risk estimation using U.S. Environmental Protection Agency standard exposure factors, and assuming that 10% of total As is the inorganic form and 100% of the total Hg is methyl Hg, predicted adverse health effects (cancer and noncancer) from fish consumption, being higher for children than for adults. However, speciation of As in fish muscle is recommended for better risk estimates. Sierra fish from Jobos Bay triggered the most restricted consumption advisories for Hg noncancer effects, where a child should not consume >1 fish meal (0.1135 kg)/month and adults should not have >3 fish meals (0.227 kg)/month. Fish consumption advisories, particularly for Hg, should be established by the local government to protect the health of susceptible populations such as children and pregnant or childbearing-age women.

Heavy metals are among the aquatic pollutants most frequently detected in estuarine and coastal areas. Despite the fact that metals occur naturally in the environment, human activities (i.e., urbanization and industrialization) can augment their natural background concentrations. Metals can enter into the food web through direct consumption of water or organisms, or through uptake processes, and be potentially accumulated in edible fish (Paquin et al. 2003). Although fish consumption provides many benefits for cardiovascular health (Burger and Gochfeld 2007), there is a growing concern that metals accumulated in fish muscle tissues may represent a higher health risk than a health benefit, especially for populations with high fish consumption rates (Liao and Ling 2003; Burger and Gochfeld 2007; Díez et al. 2009). For instance, metals like lead (Pb) and mercury (Hg) are known neurotoxicants; arsenic (As) has been associated to various systemic effects like cardiovascular disease, skin disorders, and neurotoxicity; and cadmium (Cd) is associated with nephrotoxicity (Liu et al. 2008). In addition, some metals like As and Cd have been implicated in various types of cancer (e.g., skin, bladder, lung) (Liu et al. 2008). These health concerns become of greater importance when considering susceptible populations such as young children or women of childbearing age. Recreational or subsistence fishermen (and their families) tend to frequently consume their own fish catch

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as a main source of protein, consuming much higher quantities of fish than the general population (Burger et al. 1992; Burger and Gochfeld 2007). This is the case for fishing communities like those found in the Jobos Bay area in Puerto Rico.

Jobos Bay, located in the southern part of Puerto Rico (Fig. 1), is part of the Jobos Bay National Estuarine Research Reserve (JBNERR). JBNERR contains the second largest estuarine area (Jobos Bay and adjacent areas) in Puerto Rico and, because of its high ecological importance, was designated a reserve in 1981 (DNER 2000). This reserve is economically important for marine recreation, ecotourism, commercial, and recreational fishing (DNER 2000). However, there is a concern that residential, agricultural, and industrial development in the surroundings could result in significant release of metals into the aquatic environment, as has been shown in other estuarine and coastal regions of the world (Alonso et al. 2000; Agusa et al. 2005; Fabris et al. 2006; Summers et al. 1995). The eventual accumulation of metals in edible fish tissues represents a potential health risk to people who consume them, especially to vulnerable populations such as fishing communities.

There is a lack of studies regarding metal levels in edible fish tissues from the Jobos Bay area and whether these levels represent a human health risk for fishing communities in the area. Therefore, the objectives of this study were (1) to determine levels of As, Cd, copper (Cu), Pb, Hg, selenium (Se), and zinc (Zn) in edible muscle tissues from fish commonly captured in the Jobos Bay area; (2) to compare metal levels in fish from the Jobos Bay area to fish of the same species from the La Parguera area (used as a reference site); and (3) to estimate the health risk to local fishermen and their children from ingestion of metal-contaminated fish, utilizing consumption patterns of local fishermen.

Fig. 1 Maps of Puerto Rico within the Caribbean Region and showing the location of the Jobos Bay National Estuarine Research Reserve (JBNERR) and La Parguera (LP), the reference site

Materials and Methods

Fishermen Interviews

Information on fish consumption patterns were obtained from questionnaires provided from May 2003 to January 2004 to fishermen from four main fishing areas around Jobos Bay (Playita, Las Mareas, Aguirre, and Pozuelo) and in La Parguera area (the reference site) (Fig. 1). Figure 2 presents the approximate location of Las Mareas, Aguirre, and Pozuelo. Playita (not shown on Fig. 2) is located approximately 3 km west of Las Mareas. The questionnaire, provided once to each fisherman, also collected information on fishing and sociodemographic characteristics. This study was approved by the University of Puerto Rico (UPR) Medical Sciences Campus (MSC) Institutional Review Board for Human Research Subjects Protection Office.

Fish Sampling

Fish samples were collected during May–June 2003 by research personnel with the assistance of local fishermen, and with appropriate permits from the Puerto Rico Department of Natural and Environmental Resources. The selection of sampling stations (Fig. 2) took into consideration the approximation of potential pollutant sources to the Jobos Bay area. Fish were also collected in July 2003 from the coast of La Parguera, a popular fishing and tourist area, located in southwest Puerto Rico (Fig. 1). Since the coast of La Parguera is far from agricultural and industrial development, it was used as a reference site for comparisons of metal levels in fish of the same species collected in the Jobos Bay area. Powder-free rubber gloves were worn during the sampling operation and were changed at different sampling stations. Captured fish were identified, individually placed inside plastic bags, preserved in an ice cooler, and transported to the laboratory of the Department of Environmental Health, Graduate School of Public Health, MSC-UPR, for subsequent analyses. Sampling and processing of fish followed the U.S. Environmental Protection Agency (USEPA 2000a) guidelines.

Fish Processing, Digestion, and Analysis

In the laboratory, fish were identified to the species level (Goodson 1976; Martin and Patus 1988), and length, width, and weight measurements were recorded. Fish processing and digestion were conducted as previously described by Rodríguez-Sierra and Jiménez (2002).

A total of seven metals (As, Cd, Cu, Pb, Se, Zn, and Hg) were analyzed. The analysis of As, Cd, Cu, Pb, Se, and Zn was conducted with a Perkin Elmer Atomic Absorption Spectrophotometer (AAS), Model AAnalyst 800, using modified methods of the USEPA (1992) for graphite furnace and direct aspiration flame modes. Hg was analyzed using a flow injection mercury/hydride system (Perkin Elmer; FIAS 400).

Laboratory blanks, spiked blanks, matrix spiked samples, and the standard reference material (SRM) DORM-2–Dogfish Muscle (National Research Council, Ontario, Canada) were included in each digestion batch to validate the digestion procedure. Blank verification samples were included during the AAS analysis to ensure no carryover of metals from one sample to the other. The minimum accepted correlation coefficient of the calibration curve for each metal was 0.995.

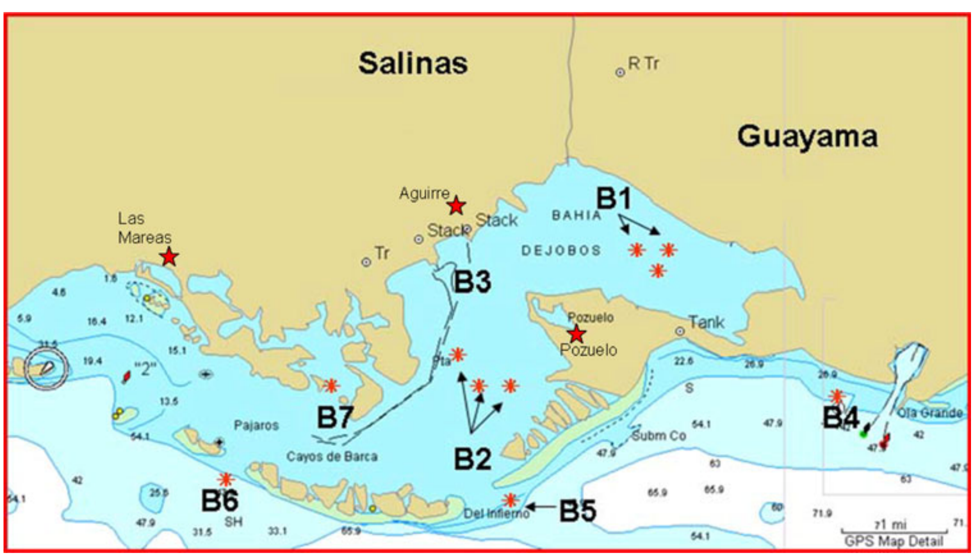

Fig. 2 Sampling stations (B1–B7) for fish in the Jobos Bay area

Statistical Analyses

Scheffe's F ($p < 0.05$) was used to examine statistical differences in average metal concentrations ($\log_{10}$ transformed) in fish muscle tissues, fish length, and fish weight between fish species and sampling sites (Jobos Bay vs. La Parguera). Statistical analyses were performed using the statistical program StatView, v. 5.0 (SAS Institute Inc., Cary, NC).

Health Risk Estimate

To estimate the human health risk from consuming metal-contaminated fish, the estimated exposure dose (EED) was calculated using the following equation (USEPA 1997):

$$\mathrm{EED} = \frac{\mathrm{Conc.} \times \mathrm{IR} \times \mathrm{EF} \times \mathrm{ED}}{\mathrm{BW} \times \mathrm{AT}}$$

where EED is the estimated metal exposure dose (mg/kg/day); Conc. is the metal concentration in fish (mg/kg, wet weight); IR is the ingestion rate (adult, 0.227 kg fish per day; child, 0.1135 kg fish per day); EF is the exposure frequency, or number of exposure events per year of exposure (from 365 days/year for people who eat fish 7 times a week to 52 days/year for people who eat fish 1 time a week); ED is the exposure duration (adult, 70 years; child, 6 years); BW is the body weight (adult, 70 kg; child, 16 kg); and AT is the averaging time or the period over which cumulative exposures are averaged ($\mathrm{AT} = \mathrm{ED} \times$ 365 days/year for noncancer risk and $\mathrm{AT} = 70$ years $\times$ 365 days/year for lifetime cancer risk). The hazard quotient (HQ) for noncancer or systemic effects was estimated dividing the calculated EED by the metal reference dose (RfD), while the lifetime cancer risk (CR) was obtained by using the chemical slope factor (SF) (USEPA 1997) in the following equation:

$$\mathrm{CR} = \mathrm{EED} \times \mathrm{SF}$$

Results

Interviews with fishermen

A total of 44 fishermen were interviewed in the four fishing areas around Jobos Bay, whereas 11 were interviewed in La Parguera. From a total of 55 fishermen interviewed, 54 were male; their average age was 54 ± 12. Results on fishing (e.g., catch record) and sociodemographic characteristics have been reported elsewhere (Mansilla-Rivera et al. 2004). Interviewed fishermen reported a total of 13 different fish species that are usually captured by them in the Jobos Bay and La Parguera areas (Fig. 3). As shown in Fig. 3, *Lutjanus synagris* ("arrayado") was the fish species most frequently reported to be captured in these areas, followed by the species *Lutjanus analis* ("sama"). *Scomberomorus* spp. ("sierra") was the fourth most frequently reported fish species.

Interviewed fishermen were asked about their fish consumption patterns. As presented in Fig. 4, 19% of respondents ($n = 53$) reported to have at least 6 fish meals/week, whereas 26% consume fish 3–5 times/week. Only 13% reported to have <1 fish meal/week. In addition, 98% of the interviewed fishermen who responded to the question about source of fish consumed ($n = 50$) reported that fish they or their families consume is caught by themselves or a close relative. These results show that the fish species included in this study are frequently captured and consumed within the fishing community of the Jobos Bay and La Parguera areas.

Fish Collection

A total of 64 specimens, comprising four different fish species, were captured from seven stations in the Jobos Bay area (Fig. 2). At stations located in the bay (B1, B2, B3, and B7), fish were captured using gill-nets; species collected were *Scomberomorus cavalla* (sierra; $n = 14$) and *Micropogon undulatus* ("roncón"; $n = 21$). In contrast, *Lutjanus synagris* (arrayado; $n = 18$) and *Lutjanus analis* (sama; $n = 11$) were captured using fish traps at stations outside the bay (B4, B5, and B6). In La Parguera, only two fish species of the same genus and/or species as those captured in the Jobos Bay area were collected: *Scomberomorus regalis* (sierra; $n = 10$) and *Lutjanus synagris* (arrayado; $n = 8$), for a total of 18 specimens. These species were captured with gill-nets and by line fishing with a hook, respectively. These two fish species, collected in the La Parguera area, were used to compare muscle–metal concentrations with *S. cavalla* and *L. synagris* trapped in the Jobos Bay area.

These fish species collected in the Jobos Bay and La Parguera areas are commonly captured by local fishermen, as shown in Fig. 3. Sierra is a pelagic top-predator fish species that occur in coastal waters and feeds mostly on clupeoid fish like sardines (Froese and Pauly 2009). Arrayado and sama are reef-associated fish that feed mainly on crustaceans and small fish, while roncón frequent mud and sandy mud bottoms in estuaries and coastal waters and feed on worms, crustaceans, and small fish (Froese and Pauly 2009).

Length and weight of fish species collected are presented in Table 1. Sierra from Jobos Bay was significantly larger (in length and weight) than the other fish species collected in the Jobos Bay and La Parguera areas. The maximum length of a sierra specimen from Jobos Bay was 67 cm, which corresponded to a weight of 1,654 g. The

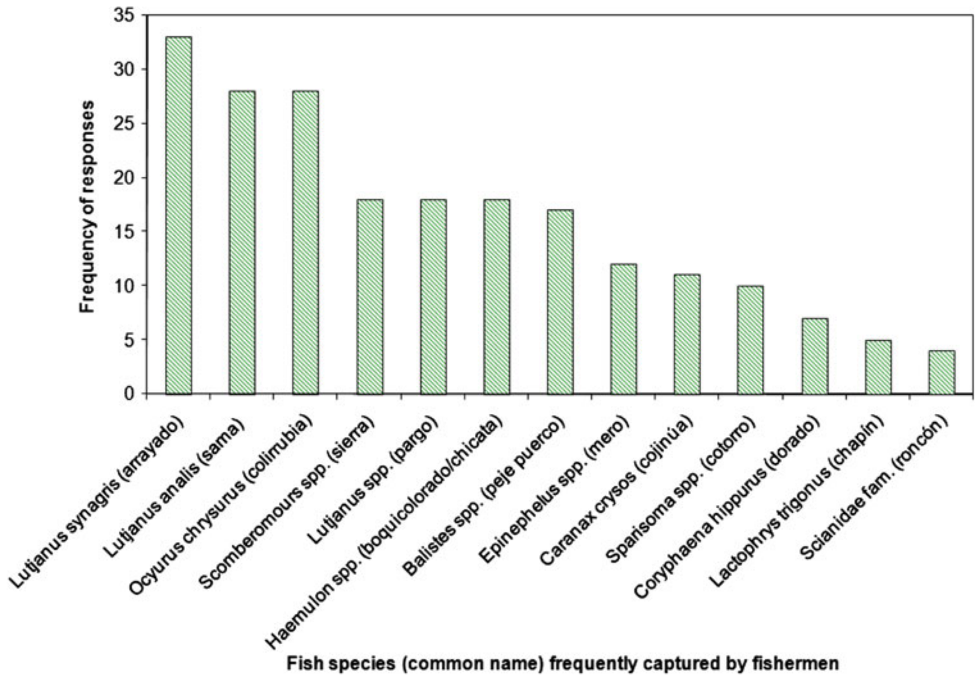

Fig. 3 Fish species frequently captured in the Jobos Bay and La Parguera areas, as reported by interviewed fishermen

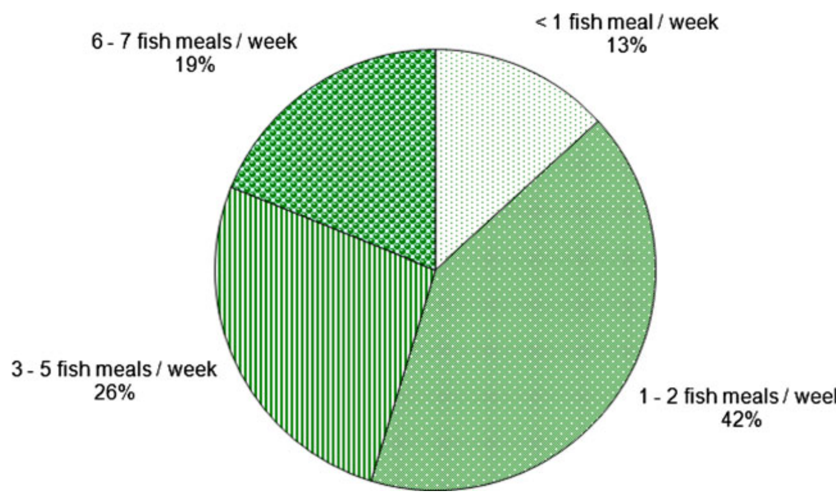

Fig. 4 Patterns of fish consumption (fish meals per week) as reported by interviewed fishermen ($n = 53$) in the Jobos Bay and La Parguera areas

trend in length and weight in Jobos Bay was as follows: sierra > roncón = sama > arrayado. In La Parguera, sierra was significantly larger (in size and weight) than arrayado. Comparing length and weight in arrayado and sierra from the Jobos Bay and La Parguera areas, no statistical differences were observed between arrayado from both sites; but sierra from Jobos Bay was statistically significantly larger than sierra from La Parguera (Table 1).

Metal Analyses

Average percentage recoveries for As, Cd, Cu, Hg, Pb, Se, and Zn analyzed from spiked digested blanks varied from 89 to 100%; and those from the SRM DORM-2 dogfish muscle ranged from 81 to 103%. Since the water content in muscle tissues of all fish species analyzed was similar (77–80%), metal levels are expressed on a wet weight (ww) basis.

Table 2 presents the average concentrations of Se, Cu, Zn, As, and Hg in fish collected from all stations in the Jobos Bay and La Parguera areas. Average Cd and Pb concentrations (not shown in Table 2) were below the minimum reporting limit (0.003 and 0.013 μg/g ww, respectively) in all fish species from the Jobos Bay and La Parguera areas. Comparisons of metal concentrations among fish species from each site showed that average levels of Se were significantly higher in roncón (0.38 μg/g ww) from the Jobos Bay area. In contrast, the average concentration of Cu was measured at significant higher levels in sierra (0.25 μg/g ww) than in the other three fish

Table 1 Average fish length and weight ± standard error of fish species collected in Jobos Bay and La Parguera areas

Species, “common name”	Length (cm)	Weight (g)
Jobos Bay		
Scomberomorus cavalla, “sierra” ($n = 14$)	55.2 ± 1.6[a,1]	1,028 ± 83.8[a,1]
	(46.0–67.0)	(604.4–1,653.9)
Lutjanus synagris, “arrayado” ($n = 18$)	25.8 ± 0.6[b,1]	227.5 ± 14.3[b,1]
	(22.0–31.0)	(148.7–356.2)
Micropogon undulates, “roncón” ($n = 21$)	33.7 ± 0.6[c]	433.4 ± 20.0[c]
	(28.0–38.5)	(284.9–625.5)
Lutjanus analis, “sama” ($n = 11$)	33.2 ± 1.8[c,d]	498.9 ± 85.7[c,d]
	(25.0–46.0)	(204.4–1,155.4)
La Parguera		
Scomberomorus regalis, sierra ($n = 10$)	41.9 ± 2.4[a,2]	525.0 ± 116.1[a,2]
	(33.0–59.0)	(202.3–1,457.5)
Lutjanus synagris, arrayado ($n = 8$)	26.2 ± 1.9[b,1]	270.9 ± 63.7[b,1]
	(20.5–35.5)	(113.7–612.0)

Note: n = number of individuals analyzed. Ranges are given in parentheses. Different superscript letters (a–d) represent statistically significant differences among species from the same site; different superscript numbers (1, 2) represent statistically significant differences between the two sites for the same fish species (sierra vs. sierra and arrayado vs. arrayado)

species from the Jobos Bay area. In the Jobos Bay area, the average level of Zn was also significantly higher in sierra (3.35 µg/g ww) than in arrayado (2.70 µg/g ww) and roncón (2.65 µg/g ww), but not significantly different from that in sama (2.83 µg/g ww). In La Parguera, average concentrations of Cu and Zn were significantly higher in sierra (0.27 and 4.01 µg/g ww, respectively) than in arrayado (0.15 and 2.58 µg/g ww, respectively), but no differences between species were observed for Se (Table 2).

In the case of As, average levels in all fish species studied ranged from 0.74 to 2.49 µg/g ww in Jobos Bay and from 0.61 to 1.27 µg/g ww in the La Parguera area (Table 2). At both sites, the arrayado was the species with the highest significant average concentration of As. Individual concentrations of As in arrayado were found to be as high as 11.72 µg/g ww in Jobos Bay and 2.97 µg/g ww in La Parguera. In the Jobos Bay area, the average As concentration in arrayado was about two- to threefold higher in comparison with the other three fish species. The same significance was observed for average As levels between arrayado and sierra from La Parguera.

Average Hg levels in the four fish species varied from 0.09 to 0.26 µg/g ww in the Jobos Bay area and from 0.12 to 0.20 µg/g ww in La Parguera (Table 2). The highest individual Hg concentration was found in sierra from Jobos Bay, at 0.40 µg/g ww. Average levels of Hg in sierra (0.26 µg/g ww) from Jobos Bay were significantly higher than in the other fish species from the same site. In contrast, in La Parguera, arrayado obtained significantly higher average concentrations of Hg (0.20 µg/g ww) than sierra (0.12 µg/g ww).

Comparing average metal levels in the same fish species (sierra and arrayado) from the Jobos Bay area versus La Parguera (used as a reference site), only Hg exhibited significantly higher concentrations in sierra from Jobos Bay (0.26 µg/g ww) than in sierra from La Parguera (0.12 µg/g ww) (Table 2).

Health Risk Estimate

Concentrations measured for Cu and Zn in all six fish species studied (Jobos Bay and La Parguera) were below the international criteria in edible fish muscle for human protection, while Se has no international criterion available (Table 2). In contrast, some levels obtained for As and Hg surpassed the international criterion established for As of 2 µg/g ww (Summers et al. 1995) and the USEPA criterion of As tissue residues in fish of 1.3 µg/g ww for human protection (Burger et al. 2007a; Burger and Gochfeld 2005) (Table 2) or were close to the Canadian standard of total Hg of 0.5 µg/g ww (HC 2007) and the more restrictive USEPA water quality criterion for methyl mercury of 0.3 µg/g fish tissue ww (USEPA 2001) (Table 2). Therefore, the potential health risk for metals in fish tissues was calculated only for As and Hg, since these two elements surpassed or were close to international and USEPA criteria used for human health protection (Table 2).

To estimate the human health risk from consuming metal-contaminated fish, the estimated exposure doses were calculated for inorganic As (assuming that 10% of the total average As in muscle tissues is the inorganic toxic form, as suggested by the U.S. Food and Drug Administration [USFDA]) and methyl Hg (assuming that 100% of the total average Hg is in the form of methyl Hg) (USFDA 1993; USEPA 2000a). The reference doses of As and Hg used to determine HQ values were 3×10^{-4} mg/kg/day (IRIS 1998) and 1×10^{-4} mg/kg/day (IRIS 2001), respectively. An oral SF of 1.50 $(\text{mg/kg/day})^{-1}$ (IRIS 1998) was used to calculate the lifetime CR in adults from consuming As-contaminated fish.

Table 2 Average metal concentrations (μg/g ww) in muscle tissues ± standard error of fish species from Jobos Bay and La Parguera areas

Species, "common name"	Se	Cu	Zn	As	Hg
Jobos Bay					
Scomberomorus cavallai, "sierra" ($n = 14$)*	$0.30 \pm 0.01^{a,1}$	$0.25 \pm 0.01^{a,1}$	$3.35 \pm 0.19^{a,1}$	$1.00 \pm 0.06^{a,1}$	$0.26 \pm 0.02^{a,1}$
	(0.25–0.34)	(0.22–0.28)	(2.28–4.51)	(0.49–1.28)	(0.18–0.40)
Lutjanus synagris, "arrayado" ($n = 18$)	$0.27 \pm 0.01^{a,1}$	$0.18 \pm 0.01^{b,1}$	$2.70 \pm 0.13^{b,1}$	$2.49 \pm 0.60^{b,1}$	$0.15 \pm 0.01^{b,1}$
	(0.17–0.39)	(0.12–0.31)	(2.15–4.56)	(0.76–11.72)	(0.08–0.25)
Micropogon undulates, "roncón" ($n = 21$)	0.38 ± 0.02^{b}	0.18 ± 0.02^{b}	2.65 ± 0.09^{b}	0.74 ± 0.13^{a}	0.10 ± 0.02^{c}
	(0.21–0.62)	(0.10–0.43)	(1.95–3.75)	(0.21–2.58)	(< mrl–0.36)
Lutjanus analis, "sama" ($n = 11$)	0.27 ± 0.02^{a}	0.15 ± 0.01^{b}	$2.83 \pm 0.13^{a,b}$	0.83 ± 0.12^{a}	$0.09 \pm 0.01^{b,c}$
	(0.17–0.39)	(0.12–0.19)	(2.08–3.42)	(0.20–1.47)	(< mrl–0.17)
La Parguera					
Scomberomorus regalis, sierra ($n = 10$)	$0.28 \pm 0.01^{a,1}$	$0.27 \pm 0.02^{a,1}$	$4.01 \pm 0.29^{a,1}$	$0.61 \pm 0.07^{a,1}$	$0.12 \pm 0.02^{a,2}$
	(0.22–0.32)	(0.19–0.39)	(2.84 ± 5.92)	(0.50–1.21)	(< mrl–0.22)
Lutjanus synagris, arrayado ($n = 8$)	$0.23 \pm 0.03^{a,1}$	$0.15 \pm 0.01^{b,1}$	$2.58 \pm 0.06^{b,1}$	$1.27 \pm 0.27^{b,1}$	$0.20 \pm 0.02^{b,1}$
	(0.04–0.34)	(0.07–0.20)	(2.37–2.92)	(0.48–2.97)	(0.17–0.26)
mrl	0.05	0.05	0.5	0.05	0.08
International criterion (Summers et al. 1995; HC 2007)	nr	15	60	2	$0.5^{\#}$
USFDA (2001) action limit					1
USEPA (2001) water quality criterion					0.3

Note: mrl, minimum reporting limit of the AAS (μg/g ww) assuming no matrix interference; nr, not reported. n = number of individuals analyzed. * $n = 13$ for the determination of Zn in sierra from Jobos Bay. Ranges are given in parentheses. Different superscript letters (a–c) represent statistically significant differences among species from the same site; different superscript numbers (1, 2) represent statistically significant differences between the two sites for the same fish species (sierra vs. sierra, arrayado vs. arrayado)

The estimates of risks to human health due to exposure to inorganic As and methyl Hg from consumption of fish are presented in Tables 3 and 4, respectively. When considering different levels of exposure (based on frequency of fish consumption) to inorganic As, the calculated HQ for children was higher than 1 for all fish species from both sites in at least one of the scenarios considered (Table 3). An HQ higher than 1 implies that the EED surpassed the USEPA reference dose for the contaminant of interest (in this case inorganic As) and that systemic effects (e.g., cardiovascular disease, skin disorders, and neurotoxicity) may occur. For adults, the EED surpassed the reference dose of 3×10^{-4} mg/kg/day when consumption of arrayado (from both Jobos Bay and La Parguera) and sierra from Jobos Bay was considered. The species that seems to represent the highest risk for systemic effects due to inorganic As exposure was arrayado from Jobos Bay: a child who eats this fish species twice a week and an adult consuming this fish species four times a week would obtain an HQ higher than 1. When estimating the lifetime excess CR for As, it can be seen that all scenarios considered obtained a CR higher than 1 in 100,000 (10^{-5}), a risk above the 10^{-6} value considered by the USEPA (1989) as an acceptable risk for cancer.

When estimating the risk due to exposure to methyl Hg from the consumption of fish (assuming that 100% of total Hg in muscle is present as methyl Hg), the calculated HQs were >1 for all fish species in Jobos Bay and La Parguera in most scenarios considered (Table 4), meaning that systemic adverse health effects (e.g., neurodevelopmental effects) may occur in the exposed population. The fish species sierra from Jobos Bay obtained the highest HQ values for children (18.4) and adults (8.4), and HQ values were >1 in all scenarios considered (even when consuming sierra from Jobos Bay once a week, HQ values for children and adults were 2.6 and 1.2, respectively). This was the fish species that obtained the highest levels of Hg in muscle, with a maximum value of 0.40 μg/g ww.

Discussion

This study determined that, from seven metals studied in muscle tissues of edible fish from the Jobos Bay and La Parguera areas in Puerto Rico, only As and Hg were detected at levels of human health concern. Average levels of As measured in fish muscles of species captured for this study (0.61–2.5 μg/g ww) were similar to those observed in three fish species (*P. dentatus*, *M. saxatilis*, *and M. undulates*) from the Mid-Atlantic region, which ranged from 0.69 to 2.61 μg/g ww (Greene and Crecelius 2006) and to average As levels found in muscle tissues of Pacific cod collected from four islands of the Aleutian Chain in Alaska (0.538–2.75 μg/g ww) (Burger et al. 2007c). In contrast, compared to As levels in tropical fish from American

Table 3 Health risk estimates for inorganic As ingestion from fish captured in the Jobos Bay and La Parguera areas

Species	Level of exposure (days/week)	Avg As conc. (mg/kg ww)	Est. inorg. As (mg/kg ww)	HQ RfD = 0.0003 mg/kg/day		Cancer risk SF = 1.5 $(mg/kg/day)^{-1}$: adult
				Adult	Child	
Jobos Bay						
Scomberomorus cavalla (sierra)	7	1.00 ± 0.06	0.1	**1.1**	**2.4**	**2.1×10^{-4}**
	5			0.8	**1.7**	**1.5×10^{-4}**
	4			0.6	**1.3**	**1.2×10^{-4}**
	2			0.3	0.7	5.9×10^{-5}
	1			0.2	0.3	3.0×10^{-5}
Lutjanus synagris (arrayado)	7	2.5 ± 0.6	0.25	**2.7**	**5.9**	5.2×10^{-4}
	5			**1.9**	**4.2**	**3.7×10^{-4}**
	4			**1.5**	**3.4**	**3.0×10^{-4}**
	2			0.8	**1.7**	**1.5×10^{-4}**
	1			0.4	0.8	**7.4×10^{-5}**
Micropogon undulatus (roncón)	7	0.74 ± 0.13	0.074	0.8	**1.7**	**1.5×10^{-4}**
	5			0.6	**1.2**	**1.1×10^{-4}**
	4			0.5	1.0	**8.8×10^{-5}**
	2			0.2	0.5	**4.4×10^{-5}**
	1			0.1	0.2	**2.2×10^{-5}**
Lutjanus analis (sama)	7	0.83 ± 0.12	0.083	0.9	**2.0**	**1.7×10^{-4}**
	5			0.6	**1.4**	**1.2×10^{-4}**
	4			0.5	**1.1**	**9.9×10^{-5}**
	2			0.3	0.6	**4.9×10^{-5}**
	1			0.1	0.3	**2.5×10^{-5}**
La Parguera						
Scomberomorus regalis (sierra)	7	0.61 ± 0.07	0.061	0.7	**1.4**	**1.3×10^{-4}**
	5			0.5	1.0	**9.1×10^{-5}**
	4			0.4	0.8	**7.2×10^{-5}**
	2			0.2	0.4	**3.6×10^{-5}**
	1			0.1	0.2	**1.8×10^{-5}**
Lutjanus synagris (arrayado)	7	1.27 ± 0.27	0.127	**1.4**	**3.0**	**2.6×10^{-4}**
	5			1.0	**2.1**	**1.9×10^{-4}**
	4			0.8	**1.7**	**1.5×10^{-4}**
	2			0.4	0.9	**7.5×10^{-5}**
	1			0.2	0.4	**3.8×10^{-5}**

Note: HQ, hazard quotient. Avg As conc.: average As concentration in fish muscle. Est. inorg. As: estimated inorganic As in fish muscle (10% of total As).Numbers in boldface represent hazard quotient values > 1 or cancer risks > 10^{-6}

Samoa (0.23–26.9 µg/g ww) (Peshut et al. 2008), results from our study were generally lower. Conversely, average As concentrations obtained in this study were much higher than those measured in striped mojarra fish (*D. plumieri*) collected in three tropical estuarine/coastal areas from Puerto Rico: San José Lagoon (0.14–0.38 µg/g ww), La Parguera (0.21 µg/g ww), and Joyuda Lagoon (0.43 µg/g ww) (Rodríguez-Sierra and Jiménez 2002). In the case of Hg, average concentrations measured in fish muscles from Jobos Bay and La Parguera (0.09–0.26 µg/g ww) were much higher than average Hg levels found in striped mojarra from San José Lagoon (0.008–0.042 µg/g ww), La Parguera (0.01 µg/g ww), and Joyuda Lagoon (0.01 µg/g ww) (Rodríguez-Sierra and Jiménez 2002). In contrast, they were similar to Hg levels measured in muscle tissues of robalo (*Centropomus* spp.) captured from lagoons and marshes at Humacao and Boquerón, Puerto Rico, where Hg concentrations ranged from 0.02 to 0.40 µg/g ww (Burger et al. 1992), and to those measured in the top predator Pacific cod (*G. macrocephalus*) from the Aleutians (0.111–0.316 µg/g ww) (Burger et al. 2007c). However, higher Hg levels have been reported elsewhere, like those found in the red drum from Florida waters, with concentrations as high as 1.70 µg/g ww (Adams and Onorato 2005).

Table 4 Health risk estimates for methyl Hg ingestion from fish captured in Jobos Bay and La Parguera areas

Species	Level of exposure (days/week)	Hg Avge Conc Est Methyl Hg (mg/kg ww)	Hazard Quotient RfD = 0.0001 mg/kg/day	
			Adult	Child
Jobos Bay				
Scomberomorus cavalla (sierra)	7	0.26 ± 0.02	**8.4**	**18.4**
	5		**6.0**	**13.1**
	4		**4.8**	**10.5**
	2		**2.4**	**5.3**
	1		**1.2**	**2.6**
Lutjanus synagris (arrayado)	7	0.15 ± 0.01	**4.9**	**10.6**
	5		**3.5**	**7.6**
	4		**2.8**	**6.1**
	2		**1.4**	**3.0**
	1		0.7	**1.5**
Micropogon undulatus (roncón)	7	0.10 ± 0.02	**3.2**	**7.1**
	5		**2.3**	**5.1**
	4		**1.8**	**4.0**
	2		0.9	**2.0**
	1		0.5	1.0
Lutjanus analis (sama)	7	0.09 ± 0.01	**2.9**	**6.4**
	5		**2.1**	**4.5**
	4		**1.7**	**3.6**
	2		0.8	**1.8**
	1		0.4	0.9
La Parguera				
Scomberomorus regalis (sierra)	7	0.12 ± 0.02	**3.9**	**8.5**
	5		**2.8**	**6.1**
	4		**2.2**	**4.9**
	2		**1.1**	**2.4**
	1		0.6	**1.2**
Lutjanus synagris (arrayado)	7	0.20 ± 0.02	**6.5**	**14.2**
	5		**4.6**	**10.1**
	4		**3.7**	**8.1**
	2		**1.8**	**4.0**
	1		0.9	**2.0**

Note: Numbers in bold represent HQ values higher than 1; Hg Avge Conc = Hg average concentration in fish muscle; Est Methyl Hg = estimated methyl mercury concentration in fish muscle (100% of total Hg)

The ongoing discussion about health benefits and risks from consumption of fish is a matter of importance, especially for communities with high fish consumption rates such as recreational or subsistence fishermen (Burger et al. 2007b). Since consumption patterns reported in this study can be classified as high (Fig. 4), our results are of great relevance. For instance, comparing consumption patterns reported in this study with those previously reported elsewhere, it is observed that they are similar to fish consumption patterns in Spain (2.5 meals/week), considered a high-fish-consumption country (Díez et al. 2009), and to those reported by subsistence people such as the Aleuts, who can eat ≥5 fish meals/week (Burger et al. 2007c). Compared to other fishermen communities in Puerto Rico, where the average fish consumption was 4.4 fish per week (average weight of each fish was 595 g) (Burger et al. 1992), consumption patterns reported in our study were lower in terms of ingestion rate (g/day) but not in terms of frequency of consumption (meals/week). In contrast, they seem higher than fish consumption patterns observed in other populations such as on the East Coast of the United States (USA), where average consumption of fish was 4.9 meals/month (Burger and Gochfeld 2009). Given the large variations observed in fish consumption patterns throughout different populations, as well as the debate on counterbalancing risks and benefits of fish consumption, it is important to consider the variation of consumption patterns when estimating potential risks associated with fish consumption. Even within the North American tribes, large variation in fish consumption rates exists, varying from 63.2 to 540 g/day (Donatuto and Harper 2008). Therefore, fish consumption surveys should be designed so they are able to capture such differences (e.g. in frequency, amount and parts of fish consumed, cooking methods, seasonal patterns of consumption), targeting specific populations that may be at higher risk due to their specific fish consumption patterns (Donatuto and Harper 2008; Moya et al. 2008). In this study, the questionnaire provided to fishermen was not specifically designed for conducting a risk assessment study; thus, parameters for calculating the exposure dose such as ingestion rate, exposure frequency, and exposure duration were only standard default exposure factors for the general population provided by the USEPA (1997). Nevertheless, the consumption pattern (i.e., exposure frequency) information provided by local fishermen aided in reducing, to a certain degree, the uncertainties of these risk estimates. For instance, after analyzing the answers provided by participants, it was shown that they frequently capture and consume the fish species included in this study (Figs. 3, 4). In addition, the assumption in this study of an average fish meal size of approximately 0.227 kg (equivalent to 8 oz) per day has been previously reported in occupational and recreational fishers (Harris et al. 2009). Therefore, the estimates used for this analysis were not that different from the real consumption patterns of local fishermen.

To protect human health, several criteria for metal levels in fish have been set. For instance, there is an international criterion for As in fish of 2 μg/g ww (Summers et al. 1995) and USEPA has set an As tissue residue of 1.3 μg/g ww in freshwater fish as the criterion for human health protection (Burger et al. 2007a; Burger and Gochfeld 2005). Likewise, USFDA has an action level for methyl Hg in fish of 1 μg/g ww (USFDA 2001). This action level, however, was not intended to be protective of populations that often capture and consume fish from the same local body of water repeatedly over many years but is only a regulatory/administrative action level (USEPA 2000b). As shown in Table 2, countries such as Canada have set lower standards for Hg in fish. In Japan, the provisional levels of total Hg and methyl Hg in fish are 0.4 and 0.3 μg/g ww, respectively (Endo et al. 2005). The USEPA promulgated in 2001 the value of 0.3 μg/g ww as a methyl Hg fish tissue residue criterion for freshwater and estuarine fish as a water quality standard (USEPA 2001). This water quality standard describes the concentration of methyl Hg in fish tissue that should not be exceeded to protect consumers of fish among the general population, based on a consumption rate of 17.5 g fish/day (USEPA 2009b). Nevertheless, to protect the health of higher fish consumption populations (e.g. subsistence fishers), screening values (SVs) with local consumption rate information and fish consumption limits should be set (USEPA 2000a, b). USEPA has calculated SVs for specific target analytes, including As and Hg, that can be used as recommended values for states when there is no specific information on consumption (e.g., for recreational fishers, SVs for inorganic As and methyl Hg are 1.2 and 0.4 μg/g, respectively) (USEPA 2000a). No SVs have been calculated for Puerto Rico.

In this study, the majority of fish individuals had metal levels below the international criterion of 2 μg/g ww for As (Summers et al. 1995) and the USEPA water quality standard of 0.3 μg/g ww for Hg in fish (USEPA 2001). For example, nine fish specimens were above the international criterion for As of 2 μg/g ww, while only three were above the water quality criteria for Hg in fish, from a total of 82 fish individuals. Nonetheless, the estimation of risk conducted in this study showed that adverse health effects may occur when considering different fish consumption patterns, as shown in Tables 3 and 4. This is why it is essential not only to report levels of contaminants in commonly captured and consumed fish species and compare them to health criteria values, but also important to establish fish consumption advisories that are easily understood by the general population.

Serious health concerns arising from consumption of contaminated fish have resulted in the establishment of various fish consumption advisories to protect human health across the USA, including most of its territories. In 2008, there were a total of 4,249 fish consumption advisories in the USA, distributed in all 50 states, the District of Columbia, five Native American tribes, and the territories of American Samoa and Guam (USEPA 2009a). However, no consumption advisories have been issued in Puerto Rico, a territory of the USA.

The methodology developed by USEPA (2000b) was applied to derive advisory consumption recommendations for minimizing the risk of both cancer and noncancer endpoints due to consumption of the fish species analyzed in this study. This methodology has also been used by other researchers to develop fish advisories for specific locations and species of fish (Huang et al. 2006; Williams and Cseh 2007; Leung et al. 2010). Metal levels used for the determination of these consumption advisories were 10% of the average total As (considered inorganic As) and the average total Hg (considered 100% methyl Hg) measured in each of the fish species.

Considering the effects of As exposure, and using the parameters suggested by USEPA (2000b), the monthly consumption limit for cancer (which represented the most

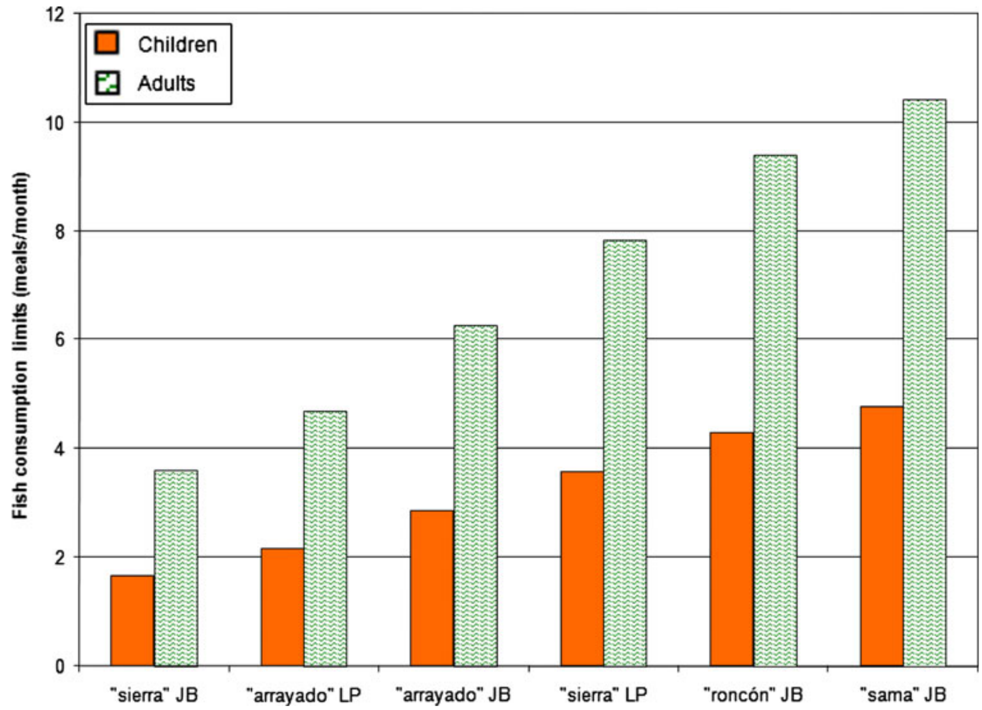

Fig. 5 Fish consumption advisories for minimizing the risk of noncancer health effects of methyl mercury in fish muscle from the Jobos Bay (JB) and La Parguera (LP) areas using USEPA (2000b) procedures. A fish meal = 0.227 kg for an adult and 0.1137 kg for a child

sensitive endpoint for As), and assuming an acceptable lifetime risk of 1 in 100,000, resulted in <1 fish meal/month for all species analyzed in this study. However, a significant limitation is that the actual percentage of inorganic As found in muscle tissues of fish species from the Jobos Bay and La Parguera areas is unknown. The assumption of having 10% of the total As in fish muscle present as inorganic As, as suggested by the USFDA (1993), is mainly based on studies conducted with fish species of temperate waters, and not with tropical fish. One of the few studies reporting inorganic As in tropical fish was conducted with tilapia fish (*O. mossambicus*) in Taiwan, in which the fraction of inorganic As was as high as 7.4% (Huang et al. 2003). This percentage was higher than others found in fish from temperate areas (<%–6.9%) (Muñoz et al. 2000; Schoof et al. 1999). Another recent study in the tropical region of American Samoa reported inorganic As in muscle tissues of four fish species ranging from below the detection limit to 4.4%, although values reported for inorganic As in whole fish were as high as 17% (Peshut et al. 2008). Moreover, it has been reported that the total As content and the species of As present in seafood may vary depending on the particular tissues that are consumed, and on how the seafood is prepared (Borak and Hosgood 2007).

Due to the uncertainty about As species present in these fish species, we feel that fish consumption advisories should not be based exclusively on levels of this element until there is more information about the distribution of As chemical forms in these fish species. Hence, average total levels of Hg (considered 100% methyl Hg, as suggested by USEPA [2000a]) were used to derive consumption limit recommendations for noncancer health effects (e.g., neurodevelopmental effects). As shown in Fig. 5, the fish species that triggered the most restricted consumption advisories for noncancer endpoints was sierra from Jobos Bay: a child should not consume more than 1 fish meal (defined as a portion of 0.1135 kg) per month, and an adult should not have sierra from Jobos Bay more than 3 times/month (fish meal = 0.227 kg). Even when considering the fish species with the lowest Hg concentration (sama from Jobos Bay), the advisories resulted in considerably stringent consumption limits (4 fish meals/month for a child and 10 for an adult). In fact, USEPA and USFDA jointly advise susceptible populations such as young children and pregnant, nursing, or childbearing-age women not to consume sierra (commonly known as king mackerel) to protect their health against harmful effects of Hg (USEPA and USFDA 2004).

The counterbalancing of health benefits and risks from fish consumption is still a critical issue that needs to be addressed, especially for communities with high fish consumption rates. Health risk estimates determined in this study were based on metal levels found in four specific fish species from the Jobos Bay area and two from La Parguera area, collected at a specific single time. Estimated risks in this study should not be extrapolated to other fish species in the studied areas or to different areas of Puerto Rico. Since fish consumption is considered a major exposure pathway to contaminants, particularly Hg, it is recommended to monitor metal levels in fish species included in this study, as well as in other edible fish species, and fish-dependent communities in Puerto Rico. To improve the estimates of risk from fish consumption, it is important to conduct As speciation analyses in edible fish from Puerto Rico. Furthermore, it is well known that fish may contain a variety of bioaccumulative organic chemical contaminants such as dioxins/furans, chlorinated pesticides, and polychlorinated biphenyls (PCBs) that are a health concern (Yim et al. 2006; Huang et al. 2006; Williams and Cseh 2007; Leung et al. 2010). In fact, a significant portion of fish advisories in effect in 2008 involved these contaminants (USEPA 2009a). Therefore, in addition to As speciation studies, other chemical organic contaminants of concern must be evaluated in edible fish from Puerto Rico to determine whether more restrictive consumption advisories are needed.

Conclusions

This is the first study in Puerto Rico reporting health risk estimates for As and Hg from consumption of locally captured fish from the Jobos Bay and La Parguera areas. Both elements in fish from these two areas represent a potential health risk for people who frequently consume these fish species. However, speciation of As is recommended for better risk estimates. Fish consumption advisories for these fish species from the Jobos Bay and La Parguera areas should be established by Puerto Rico government health agencies, particularly for Hg, to protect the health of susceptible populations such as young children and pregnant or childbearing-age women.

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