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Mercury concentrations in seafood and the associated risk in women with high fish consumption from coastal villages of Sonora, Mexico

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A B S T R A C T

Mercury concentrations in the ocean have increased considerably since the industrial revolution and will continue to increase in the next 50 years. Therefore, it is important to monitor Hg levels in fish and to evaluate the health risks in populations with high fish consumption. In the present study, a total of 238 samples of commercial fish and shellfish, were analyzed from the Central Gulf of California, Mexico. Concentrations of total Hg in fish ranged from < DL (detection limit) up to 1.22 μg g-1, with a mean of 0.15 ± 0.19 μg g-1, the majority of the samples were lower than the maximum permissible level. To evaluate the risk, a total of 110 food frequency questionnaires were applied to women (16–68 years old) from 15 coastal fishing villages of Sonora. Results indicated a high seafood consumption at these communities (mean = 307 g day-1), and a high hazard risk (HQ = 6.2) due to methyl mercury ingestion. It is recommended to limit seafood consumption in pregnant women to 4 portions of fish per week, preferably of low mercury concentrations so that all the benefits of seafood consumption are obtained without the negative health effects of methyl mercury.

1. Introduction

Mercury in the upper ocean has tripled since the beginning of the Industrial Revolution due to human activities (Lamborg et al., 2014). The circulation patterns that move cold, salty and dense water to the ocean depths also transport mercury from shallower depths; this natural process of mercury sequestration may soon become limited as increasing human inputs have been predicted to involve as much mercury in the next 50 years as in the previous 150 years (Lamborg et al., 2014). In addition, mercury previously released from human activities continues to cycle through the atmosphere, ocean and terrestrial systems for hundreds of years, constituting two thirds of total annual emissions to the atmosphere (Pirrone et al. 1996, 2009; Sunderland and Selin, 2013). Mercury is emitted to the atmosphere as inorganic Hg°, which is highly volatile, but it can be removed after oxidation to Hg(II) and deposition onto land and ocean (Boening, 2000). This species of Hg is then transformed to organometallic compounds such as methyl mercury (MeHg) under anaerobic conditions or by mercury methylating microorganisms (Jackson, 1991). Bacterial methylation process can also occur inside fish intestines, as was proven by Rudd et al. (1980). Measurements of dissolved gaseous mercury (DGM) made over the Pacific Ocean reported elevated concentrations of methylated Hg species present in the low oxygen waters of the equatorial Pacific, both MeHg and Me_2Hg were identified and concentration and distribution reflected to some degree surface ocean productivity (Kim and Fitzgerald, 1986; Mason and Fitzgerald, 1990, 1991; Macdonald et al., 2008; Sprovieri et al., 2009). This methylated form of Hg is highly lipophilic and consequently accumulates in fatty tissues of marine organisms and biomagnifies in the food chain (Hall et al., 1997; Bodalay and Fudge, 1999; Dietz et al., 2000; Dabeka et al., 2011; Copat et al., 2014; Conte et al., 2015; Adel et al., 2016; Ferrante et al., 2017). According to the U.S.EPA (2011), Revised Risk Assessment of Hg the dominant human exposure pathway is through the consumption of fish that have bioaccumulated Hg and the main health effect is neurological deficits in children who were exposed to MeHg *in utero* primarily through maternal fish consumption (Davidson et al., 1996; Copat et al., 2014; Carocci et al., 2014; Llop et al., 2015).

It is well established that fish from higher trophic levels, accumulate more mercury (Penedo de Pinho et al., 2002; Cizdziel et al., 2002; Sampaio da Silva et al., 2005; Dabeka et al., 2011). In the Gulf of California, sharks (i.e. smooth hammerhead, pelagic thrasher, pacific sharpnose shark, dusky shark, scalloped hammerhead and whitenose shark) have been identified as having high mercury concentrations (> 0.5 ppm total mercury or “THg”) in the Gulf of California (García-Hernández et al., 2007; Ruelas-Inzunza et al., 2008; Boucher, 2013; Bergés-Tiznado et al., 2015). However, the fishery of sharks and other

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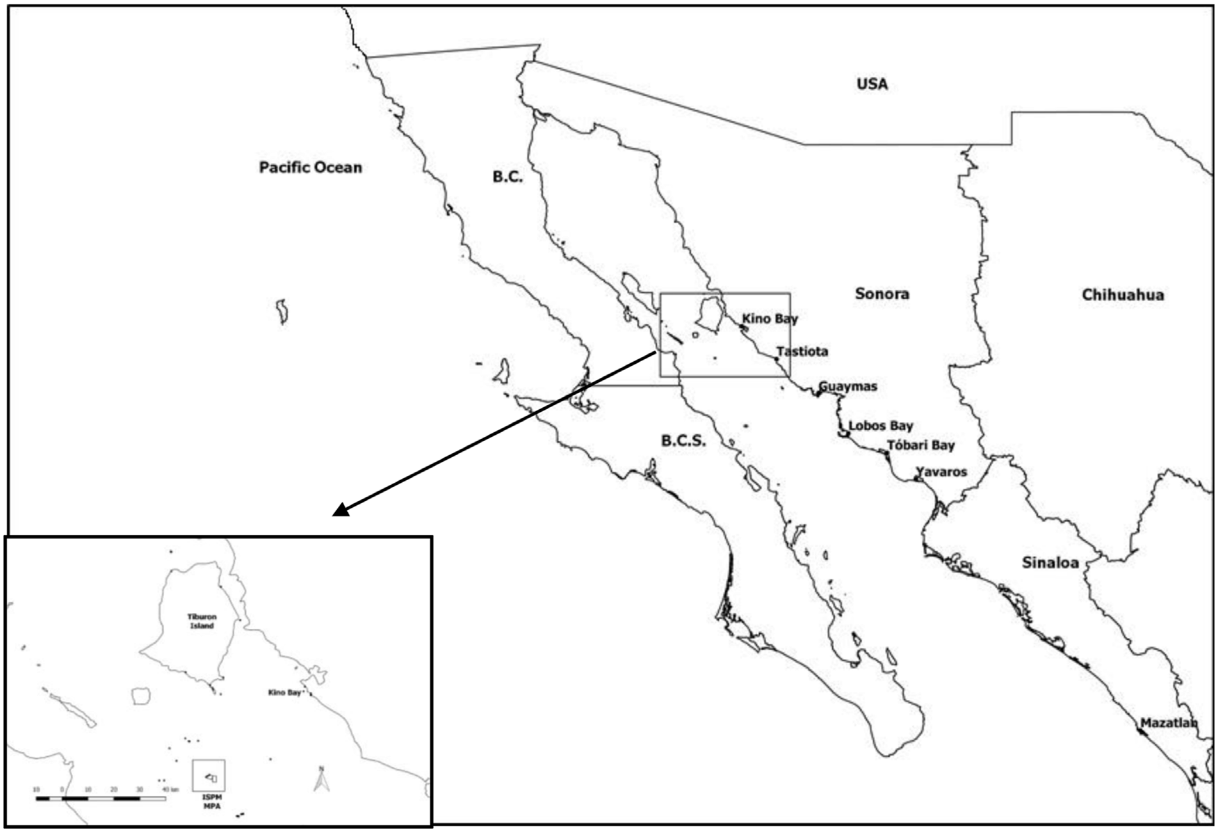

Fig. 1. Location of fish sampling points, and coastal regions where food frequency surveys were conducted in 2012.

predator species represent only 7% of all fish species harvested in Mexico (excluding small pelagic fish) (CONAPESCA, 2014). Some of these more highly consumed species are tuna, groupers, snappers, mullets, corvine, sole, tilapia, among others (CONAPESCA, 2014). Ruelas-Inzunza et al. (2011a) reported higher concentrations of mercury in piscivorous fish from the coasts of Sonora and Sinaloa compared with omnivores or herbivores fish, although concentrations not exceeded the maximum permissible levels (MPL). Authors also estimated a methyl mercury hazard quotient (HQ) for these species, considering an average national fish consumption rate of 25 g day^{-1}, concluding that no risk was identified for these species, although, they recognized a greater potential risk in fishermen and their families due to higher fish consumption.

The Gulf of California contains world-ranked, high marine biodiversity, with ca. 6000 species of invertebrates, sharks, rays, fish, sea turtles, aquatic birds and mammals (Brusca et al., 2005). The Midriff Islands region, in the northern Gulf of California (Fig. 1), is a national key site for the industrial (small pelagic fish, shrimp) as well as small-scale and sport fisheries (Kira, 1999; Cisneros-Mata, 2010; Erisman et al., 2011). The small-scale and sport fisheries capture up to 70 different species (Moreno-Báez et al., 2012), of which an important portion are captured in the coastal and deep rocky reefs along the Baja California and Sonora states coasts and around the 45 islands and islets of the region. Studies indicate that mercury concentrations are not elevated at the Midriff Region of the Gulf of California in different biota (Cahill et al., 1998; García-Rico and Ramos-Ruiz, 2001; García-Hernández et al., 2015; Páez-Osuna et al., 2017). Therefore, the levels of Hg in the Midriff Region of the Gulf of California, from a human-food viewpoint, would be mostly below hazardous levels for human consumption. However, these levels will not stay the same in the future and conclusions relating the general population might not be the same for fishermen and their families who consume fish more regularly.

In this context, the three main objectives of the present study were: 1) to determine mercury concentrations in various commercial species of fish and shellfish from the central Gulf of California; 2) to describe the food consumption patterns of women from fishing villages of central and southern Sonora; and 3) to estimate the health hazards associated to dietary intake of methyl mercury in women from these communities.

2. Materials and methods

2.1. Fish and shellfish collections

Collection of organisms were made with two efforts. The first one was from February 2008 to June 2009 at the central-east portion of the Gulf of California (Fig. 1). Sampling points were located at the proximity of Kino Bay, Tiburon Island and outside the boundaries of Isla San Pedro Martir Biosphere Reserve (ISPM-BR) (Fig. 1). Collection of fish were made by hook and line onboard a small-scale fishery boat. Species were identified, measured (total length) to the nearest mm and weighted to the nearest gram. A portion of muscle of approximately 2 cm^2 was extracted from the left side of the dorsal fin and saved in a clean resealable plastic bag. Due to the high number of fish collected for some of the species, tissues corresponding to five fish of the same species and same size class, were homogenized into one pooled composite sample (Table 1). A second effort corresponded to the years 2012, 2015, 2016 and 2017 at Kino and Tobari Bays (Table 1, Fig. 1). Organisms were collected by trained fishermen, who identified, measured, weighed and collected a muscle tissue sample following the same methodology as the first sampling survey.

2.2. Tissue analysis

At the laboratory, samples were digested using a microwave oven (CEM Corp. $MARS_X$, Matthews NC, U.S.A). Approximately 1 g of fresh

Table 1
Mean, 75th percentile and range of Hg concentration (μg g^{-1} wet wt.) in fish and shellfish species from the Gulf of California. Number of samples and number of composite samples in parenthesis.

Species	Common name[a]	N	Mean Hg conc.	75th percentile Hg conc.	Range Hg conc.	Collection year(s)/ comments
Fish						
1) *Seriola lalandi*	yellowtail jack	15 (3)	0.113	0.702	0.06–0.702	2012
2) *Dasyatis brevis*	whiptail stingray	27 (12)	0.390	0.675	0.09–1.00	2012, 2015, 2016, 2017
3) *Hoplopagrus guentherii*	barred pargo	12	0.218	0.321	0.038–0.508	2012, 2015, 2016, 2017
4) *Paralichthys aestuarius*	Cortez halibut	15 (6)	0.166	0.304	0.027–0.411	2012, 2015, 2016, 2017
5) *Mycteroperca jordani*	Gulf grouper	2	0.190	0.279	0.10–0.28	2008
6) *Mugil cephalus*	striped mullet	7	0.090	0.248	< DL-0.256	2012, 2015, 2016, 2017
7) *Mycteroperca rosacea*	leopard grouper	55	0.148	0.222	< DL-0.466	2008
8) *Hyporthodus niphobles*	star-studded grouper	7	0.170	0.217	< DL-0.47	2008
9) *Scarus perrico*	bumphead parrotfish	3	0.106	0.212	< DL-0.212	2008
10) *Paralabrax auroguttatus*	goldspotted bass	125 (55)	0.146	0.166	< DL-1.22	2008
11) *Caulolatilus affinis*	Pacific golden-eyed tilefish	1	0.164	0.164	–	2008
12) *Mustelus californicus*	gray smoothhound	15 (5)	0.085	0.103	0.06–0.11	2012
13) *Caulolatilus princeps*	ocean whitefish	62 (19)	0.029	0.054	< DL-0.122	2008
14) *Paralabrax maculotofasciatus*	spotted sand bass	1	< DL	< DL	–	2008
15) *Cynoscion xanthulus*	orangemouth corvina	3	< DL	< DL	–	2008
16) *Cynoscion albus*	queen corvina	3	< DL	< DL	–	2008
Bivalves						
17) *Dosinia ponderosa*	dosinia clam	36 (15)	0.169	0.147	< DL-1.12	2012, 2015, 2016, 2017
Crabs						
18) *Callinectes bellicosus*	warrior swimcrab	10	0.155	0.175	0.065–0.400	2012, 2015, 2016, 2017
Shrimp						
19) *Litopenaeus stylirostris*	blue shrimp	19	0.17	0.276	0.032–0.843	2012, 2015, 2016, 2017
Organisms not collected[b]						
Crassostrea gigas	giant oyster	40	0.03	0.03		García-Rico and Ramos-Ruiz, 2001
Various spp.	octopus and squids	45	0.056	0.056		Ahmad et al., 2015
Rimapenaeus similis, Farfantepenaeus californiensis, Sicyonia penicillata	yellow roughneck shrimp, yellowleg shrimp, target rock shrimp			0.276		concentration from (19)
Atrina maura, Hexaplex erythrostomus,	maura pen shell and pink-mouthed murex			0.147		concentration from (17)
Panulirus gracilis	green spiny lobster			0.056		concentration from (18)
Trachinotus paitensis, Trachinotus rhodopus	paloma pompano, gafftopsail pompano			0.702		concentration from (1)
Spheroides annulatus, Balistes polylepis, Bagre panamensis, Eucinostomus argenteus, Pomacanthus zonipectus	bullseye puffer, finescale triggerfish, chihuil, spotfin mojarra, Cortez angelfish			0.156		Mean concentration from (10,14,9 and 6)

< DL: under detection limit.
[a] Fish common names based on Page et al. (2013).
[b] List of fish and shellfish not collected in the present study, values were calculated from similar species analyzed and from the literature.

homogenized tissue was weighed in a HP-500 vessel with 5 ml of 50% nitric acid, the vessels were mounted in the turntable and digested for 20 min with the following program: 5 min ramp at 100 °C and 65 PSI, 5 min ramp at 120 °C and 100 PSI and 15 min hold at 140 °C and 140 PSI. A second digestion included adding 3 ml of 30% hydrogen peroxide to the sample and digesting again following the same program. Each sample was dissolved to 50 ml total volume with distilled water prior analysis (U.S.EPA, 1996). Digested samples were analyzed by hydride generation atomic absorption spectrophotometry using stannous chloride as a reducing agent (Perkin-Elmer 1100-B and MHS-20, Waltham MA U.S.A.) following EPA method 7471A (U.S.EPA, 1994). All results were expressed as μg g^{-1} (ppm) wet weight.

For Quality Control/Quality Assurance (QA/QC) purposes, blanks, duplicates and reference material (DORM-2 dogfish muscle reference certified material for trace metals, from the National Research Council Canada, NRCC) were analyzed. QA/QC results indicated a relative percent difference of 8.77 with variations between 0 and 23 and a mean percent recovery of 101 with variations between 98 and 114. The method detection limit based on spiked samples (MDL_s) (U.S EPA, 2016) corresponded to 0.004 μg g^{-1}.

2.3. Food frequency surveys

A pilot study was conducted aimed to determine mean consumption and preparation of common seafood products among adult women from the community of Kino Bay, Sonora (Fig. 1). Data showed as much as 52 species of marine organisms prepared in 170 recipes, including raw, steamed, breaded, fried and salted. To estimate the women's mean intake per each type of fish and seafood, we used a booklet with real portion-size photographs and serving utensils such as plates and spoons. This information was used to design a food frequency questionnaire (FFQ) using the 170 fish and shellfish preparations, mean portion sizes, as well as women age and weight. The FFQ included consumption frequency options (daily, weekly, monthly, annually or rarely), and portion sizes (small, mean, or large). The questionnaire also included blank rows to complete the list with seafood products consumed and not previously included in the questionnaire. Daily seafood intake was calculated using portion size and consumption frequency. The sample consisted in 110 women, ages16 to 68, from 15 small (< 6000 inhabitants) fishing villages along the coast of Sonora, Mexico (Fig. 1). Data was collected during the summer season of 2012.

2.4. Risk estimation

The risk of mercury exposure was calculated based on the U.S.EPA (2011) National-scale assessment of mercury risk to populations with high consumption of self-caught freshwater fish, report. First, the 75th percentile of mercury concentration (*HgTC*) (μg g^{-1} wet wt. of total Hg) was generated for the different species of marine organisms collected in the surveys, for species that were consumed but were not collected, an

Table 2
Concentrations of THg (μg g^{-1} wet wt.) in different species of fish and shellfish from the Gulf of California and other world regions.

Family	Species	Common name	Mean THg concentration	Location	Author
Serranidae	*Various species*	grouper	0.36	Canada	Dabeka et al., 2011
	Various species	grouper	0.09	Taiwan	Chen and Chen 2006
	Various species	grouper	0.45	U.S.A.	FDA 2016
	Mycteroperca jordani	Gulf grouper	0.36	northern gulf of CA, Mex	García-Hernández et al., 2007
	Mycteroperca jordani	Gulf grouper	0.19	central Gulf of CA, Mex	this study
	Various species	sea bass	0.33	Canada	Dabeka et al., 2011
	Various species	sea bass	0.07	UK	Knowles et al., 2011
	Various species	sea bass	0.15	U.S.A.	FDA 2016
	Paralabrax nebulifer	barred sand bass	0.22	southern California bight, U.S.A.	Phillips et al., 1997
	Paralabrax auroguttatus	goldspotted bass	0.17	northern Gulf of CA, Mex.	García-Hernández et al., 2007
	Paralabrax auroguttatus	goldspotted sand Bass	0.12	central Gulf of CA, Mex	this study
Lutjanidae	*Lutjanus colorado*	colorado snapper	0.13	Sinaloa coast, Mex	Ruelas-Inzunza et al., 2008
	Lutjanus sp.	red snapper	0.15	Canada	Dabeka et al., 2011
	Lutjanus sp.	snapper	0.11	U.S.	FDA 2016
	Chrysophrys auratus	snapper	0.32	Australia	Chvojka et al., 1990
	Lutjanus argentimaculatus	red snapper	0.08	Selangor, Malaysia	Jeevanaraj et al., 2016
	Hoplopagrus guentherii	barred pargo	0.46	central gulf of CA	this study
Scianidae	*Cynoscion xanthulus*	orangemouth corvina	0.05	Sinaloa coast, Mex	Ruelas-Inzunza and Páez-Osuna, 2005
	Cynoscion regalis	weakfish	0.24	U.S.A.	FDA, 2016
	Isopisthus remifer	bigeye corvina	0.14	Estern Pacific, Mex	Spanopoulos-Zarco et al., 2014
	Cynoscion albus	queen corvina	< DL	central Gulf of CA	this study
Malacanthidae	*Caulolatilus princeps*	ocean whitefish	0.12	Sinaloa coast, Mex	Ruelas-Inzunza et al., 2008
	Various spp.	tilefish	0.60	Canada	Dabeka et al., 2011
	Various spp.	tilefish	0.14	Atlantic ocean	FDA, 2016
	Caulolatilus princeps	ocean whitefish	0.03	central Gulf of CA	this study
Dasyatidae	*Dasyatis brevis*	whiptail stingray	0.45	northern Gulf of CA	García-Hernández et al., 2007
	Dasyatis dipterura	diamond stingray	0.21	Sonora coast	Ruelas Inzunza et al., 2013
	Dasyatis dipterura	diamond stingray	0.64	Nayarit coast	Ruelas Inzunza et al., 2013
	Dasyatis zugei	pale-edged stingray	0.09	Selangor, Malaysia	Jeevanaraj et al., 2016
	Various spp.	skate	0.13	U.S.A.	FDA, 2016
	Dasyatis brevis	whiptail stingray	0.39	central Gulf of CA	this study
Carangidae	*Seriola dorsalis*	yellowtail	0.21	Japan	Nakagawa et al., 1997
	Seriola lalandi	yellowtail jack	0.17	central Gulf of CA	Ruelas Inzunza and Páez-Osuna, 2005
	Hemicaranx leucurus	yellowfin jack	0.04	Eastern Pacific	Spanopoulos-Zarco et al., 2014
	Caranx caninus	Pacific crevalle jack	0.04	Sinaloa coast	Ruelas Inzunza et al., 2011a
	Seriola lalandi	yellowtail jack	0.11	central Gulf of CA	this study
Paralichthyidae	*Paralichthys woolmani*	dappled flounder	0.15	Sinaloa coast	Ruelas-Inzunza et al., 2008
	Cyclopsetta querna	toothed flounder	0.06	eastern Pacific	Spanopoulos-Zarco et al., 2014
	Paralichthys aestuarius	Cortez halibut	0.16	central Gulf of CA	this study
Mugilidae	*Agonostomus monticola*	mountain mullet	0.08	Sinaloa coast	Ruelas-Inzunza et al., 2011a
	Mugil cephalus	striped pullet	0.06	Selangor, Malaysia	Jeevanaraj et al., 2016
	Mugil cephalus	striped mullet	0.09	Central Gulf of CA	This study
Bivalves	Perna perna	brown mussel	0.06	Gahana	Joiris et al., 2000
	Crassostrea corteziensis	Cortez oyster	0.09	Central Gulf of CA	Jara-Marini et al., 2013
	Dosinia ponderosa	dosinia clam	0.17	Central Gulf of CA	This study
Shrimp	*Litopenaeus stylirostris*	blue shrimp	0.12	Sinaloa coast	Ruelas-Inzunza et al., 2004
	Litopenaeus stylirostris	blue shrimp	0.04	New Caledonia	Chouvelon et al., 2009
	Various spp.	shrimp	0.01	U.S.A.	FDA, 2016
	Litopenaeus stylirostris	blue shrimp	0.17	Central Gulf of CA	This study
Crabs	*Callinectes sapidus*	blue crab	0.08	Florida Atlantic coast	Adams and Engel, 2014
	Portunus segnis	blue crab	0.48	Persian Gulf	Ghaeni et al., 2015
	Callinectes bellicosus	warrior swimcrab	0.15	Central Gulf of CA	This study

estimated value based on similar species collected in this study was calculated or obtained from the literature (Table 1). Total mercury concentrations were converted to MeHg using a mercury conversion factor (*MCF*). For fish, this factor was 0.95 (unitless), since it is recognized that more than 90% of Hg in fish is MeHg (U.S.EPA, 1997; Moon et al., 2011; Ruelas-Inzunza et al., 2011b). However, for other groups of marine organisms, MeHg proportions of total Hg are much lower; the *MCF* for crustaceans and cephalopods was 0.5 and for bivalves it was 0.2, based on results by Moon et al. (2011). According to several studies, food preparation/cooking affects the final concentration of mercury in fish, these values can range from zero or even reduction of concentration (Perelló et al., 2008) up to 50% increases (Morgan et al., 1997). Using reference values for marine fish (Perelló et al., 2008; Ouédraogo and Amyot, 2011), an average preparation/cooking adjustment factor (*FPCAF*) of 1.13 (i.e. 13% increase in the MeHg concentration per unit fish) was used.

The seafood consumption rate in g day^{-1} (*SCR*) was obtained from the FFQ, as the grams per day consumed by each person of a specific seafood product (i.e. stewed shrimp, breaded sand bass, etc), the SCR_{total} was the sum of all the grams day^{-1} of the different seafood consumed by each person and the SCR_{mean} was the average of the individual seafood consumption. Body weight (*BW*, kg) was obtained only from one third of the females interviewed; therefore, a mean body weight value (72.30 kg) was used for all the calculations. These factors were combined in the following equation, to obtain the daily methyl mercury intake (*IR*) in μg kg^{-1} day (U.S.EPA, 2011):

$$IR = \frac{HgTC \times MCF \times FPCAF \times SCR}{BW}$$

An *IR* was calculated for each seafood product consumed by a person, the sum of all *IR* for each person was the total IR (IR_{total}) and the mean IR (IR_{mean}) was the average of the individual IR_{total}.

The risk was obtained with a hazard quotient (*HQ*):

$$HQ = \frac{IR}{RfD}$$

An *HQ* was calculated for each seafood product consumed by a person, the sum of all HQ for each person was the total HQ (HQ_{total}) and the mean HQ (HQ_{mean}) was the average of the individual HQ_{total}.

The reference dose (*RfD*) for MeHg used in this analysis was 0.0001 mg kg^{-1} day (equivalent to 0.1 μg g^{-1} day), this value was obtained from different studies including the health effects of mercury in populations from the Seychelles and Faroes Islands published by the U.S. EPA in the Integrated Risk Information System in 2001 (U.S.EPA, 2001). Therefore, exposures above the *RfD* (i.e. $HQ > 1$) represent a potential public health hazard and recommendations can be issued to reduce risk, especially in women at reproductive age.

3. Results and discussion

3.1. Concentrations of Hg in fish and shellfish

Total mercury concentrations in fish ranged from < DL (under detection limit) to 1.22 μg g^{-1}, with a mean of 0.15 ± 0.19 μg g^{-1} (Table 1). Only 6% of the samples exceeded the maximum permissible level (MPL) of 0.5 μg g^{-1} methyl mercury (DOF, 2011), these samples were goldspotted sand bass, yellowtail jack and whiptail stingray. The species with the lowest mean Hg concentrations were queen corvina, orangemouth corvina and spotted sand bass (Table 1). It is important to note that for some of the fish species there was a very low sample size, therefore, these values should be considered as individual concentrations, not representative of the population.

Concentrations of Hg in shellfish varied from < DL to 1.12 μg g^{-1} (Table 1), with a mean of 0.16 ± 0.22 μg g^{-1}. Higher concentrations in clams (1.12 μg g^{-1}) and shrimps (up to 0.84 μg g^{-1}) were recorded in the 2017 sampling period at the south Sonora location; 6% of the samples exceeded the MPL (DOF, 2011).

Comparisons of mercury concentrations in fish and shellfish from other regions of Mexico and the world are shown in Table 2. For Serranidae, concentrations appeared to be lower than values reported from the northern Gulf of California, the U.S.A. and Canada (Phillips et al., 1997; García-Hernández et al., 2007; Dabeka et al., 2011; FDA, 2016). For Lutjanidae, concentrations were higher than those found at the Sinaloa coast, the U.S.A., Australia and Malysia (Chvojka et al., 1990; Ruelas-Inzunza et al., 2008; Dabeka et al., 2011; FDA, 2016; Jeevanaraj et al., 2016). Weakfish and Tilefish presented the lowest concentrations compared with values from the U.S.A. and Canada (Dabeka et al., 2011; FDA, 2016). Stingrays appear to have higher mercury concentrations than the rest of the fish species, the highest concentration was reported for the Nayarit coast, located south from Sinaloa state (Ruelas Inzunza et al., 2013). According to Lambertsson et al. (2001) a source of methyl mercury is found in the top 3 cm of marine sediments, where methylation and de-methylation reactions take place by sulfate reducing bacteria. Therefore, the availability of MeHg to the benthic food-chain increase the concentrations in fish like rays, mullets and flounders, as well as invertebrates like shrimps and crabs, which are dependent on benthic organisms and algae.

Hg concentrations from yellowtail jack were comparable with previous investigations from the central Gulf of California (Ruelas-Inzunza and Páez-Osuna, 2005). Concentrations of Hg in yellowtail jacks from the Tokyo market, in Japan had a mean concentration of 0.21 μg g^{-1}, however, young yellowtail had higher Hg concentrations (0.06–0.76 μg g^{-1}) than mature fish (0.20–0.22) (Nakagawa et al., 1997). In the present study, concentrations of Hg in yellowtail jack had significant variations (from 0.06 to 0.70 μg g^{-1}); lower concentrations (0.11 μg g^{-1}) corresponded to larger individuals (average of 56 cm and 3.1 kg), while higher concentrations (0.70 μg g^{-1}) corresponded to smaller individuals (average of 44 cm and 2.5 kg). It is possible that younger fish had higher Hg concentrations; however, a larger sample size is needed to prove this hypothesis.

Concentrations in flounder were higher than those reported for the Mexican eastern Pacific (Spanopoulos-Zarco et al., 2014). And mean concentrations of mercury in mullets appear to be lower than the rest of the species, with values similar to those reported for Sinaloa (Ruelas-Inzunza et al., 2011a). Concentrations of Hg in shrimp were generally low (0.05 ± 0.01 μg g^{-1}), before the last 2017 sampling which increased to 0.49 ± 0.25 μg g^{-1}, in samples collected at the south of Sonora coast. The cause of this increase was not clearly identified, although satellite images (LANDSAT 8, GloVis) (USGS, 2018), showed tide flooding of the coastal plain, close to the agriculture and aquaculture areas, which could wash out contaminants and increase Hg levels at the coast.

Dosinia clams concentrations seem to be elevated compared with other bivalves from the Gulf of California (Jara-Marini et al., 2013; García-Rico and Ramos-Ruiz, 2001), and concentrations in crabs were higher than values reported for the Atlantic coast (FDA, 2016).

A multiple linear regression was calculated using JMP 8 (SAS Institute), to predict THg concentrations based on total length (TL), weight (W) and location (near, or off the coastline) for goldspotted sand bass and leopard grouper and ocean whitefish. A significant regression equation was found ($F_{3,51} = 20.58$, $p < 0.0001$), with an R^2 of 0.55 for goldspotted sand bass. THg concentration increased 0.0002 μg g^{-1} for each g of weight, decreased 0.007 μg g^{-1} for each cm of TL and location near the coastline had 0.08 μg g^{-1} more THg than off the coastline. Weight and location were the two significant predictors of THg concentrations for this species. For leopard grouper, no significant regression was found ($F_{3,38} = 0.66$, $p = 0.57$). For ocean whitefish, the two independent variables used were W and TL. A significant regression equation was found ($F_{2,16} = 32.6$, $p < 0.0001$), with an R^2 of 0.80. THg concentration increased 0.0001 μg g^{-1} for each g of weight and decreased 0.0006 μg g^{-1} for each cm of TL. Weight was the only significant predictor of THg for this species.

Many authors have found a correlation between THg and weight or length in marine and freshwater fish, with the general conclusion that mercury accumulates with fish age, and parameters of length and weight are good approximations of age in most species (Andersen and Depledge, 1997; Davis et al., 2002; Greenfield et al., 2005; Garcia-Hernández et al., 2007; Pouilly et al., 2012; Kehrig et al., 2013). Results from studies of diet composition and food web structure in the Gulf of California classify the goldspotted sand bass as member of the pelagic guild with their main diet consisting on shrimp, jellyfish, hake, crabs and lobsters for adult organisms, and on small deep sea and reef fish for juveniles (Ainsworth et al., 2011). This could explain the lower concentrations of THg found in smaller organisms (juveniles).

3.2. Seafood consumption

Seafood consumption rate (SCR_{mean}) among women from fishing communities of Sonora averaged 307 ± 325 g day^{-1}. Significant differences were found between the five age groups (one-way ANOVA *p-value* = 0.0079). Multiple comparisons between the different age groups identified the SCR_{mean} of women between 40 and 49 years old, as the highest consumption rate (471 g day^{-1}) compared with the group 50 or more (173 g day^{-1}) and the 20–29 age group (215 g day^{-1}). No differences were found with groups 16–19 (469 g day^{-1}) and 30–39 (297 g day^{-1}) (Tukey-Kramer HSD, alpha = 0.05) (Fig. 2).

According to the Fishery Secretary (CONAPESCA, 2014), the per capita consumption of fish and shellfish by the Mexican population is 32 g day^{-1}. However, fish and shellfish consumption at coastal communities of Sonora were almost ten times as higher the national average. High frequency of fish consumption at these communities is mainly due to the availability and affordability of fishery products, since other sources of protein like meat or poultry are expensive and not as available as fish and shellfish. High fish consumption (384 g day^{-1} for women) was observed in a fishing related population from

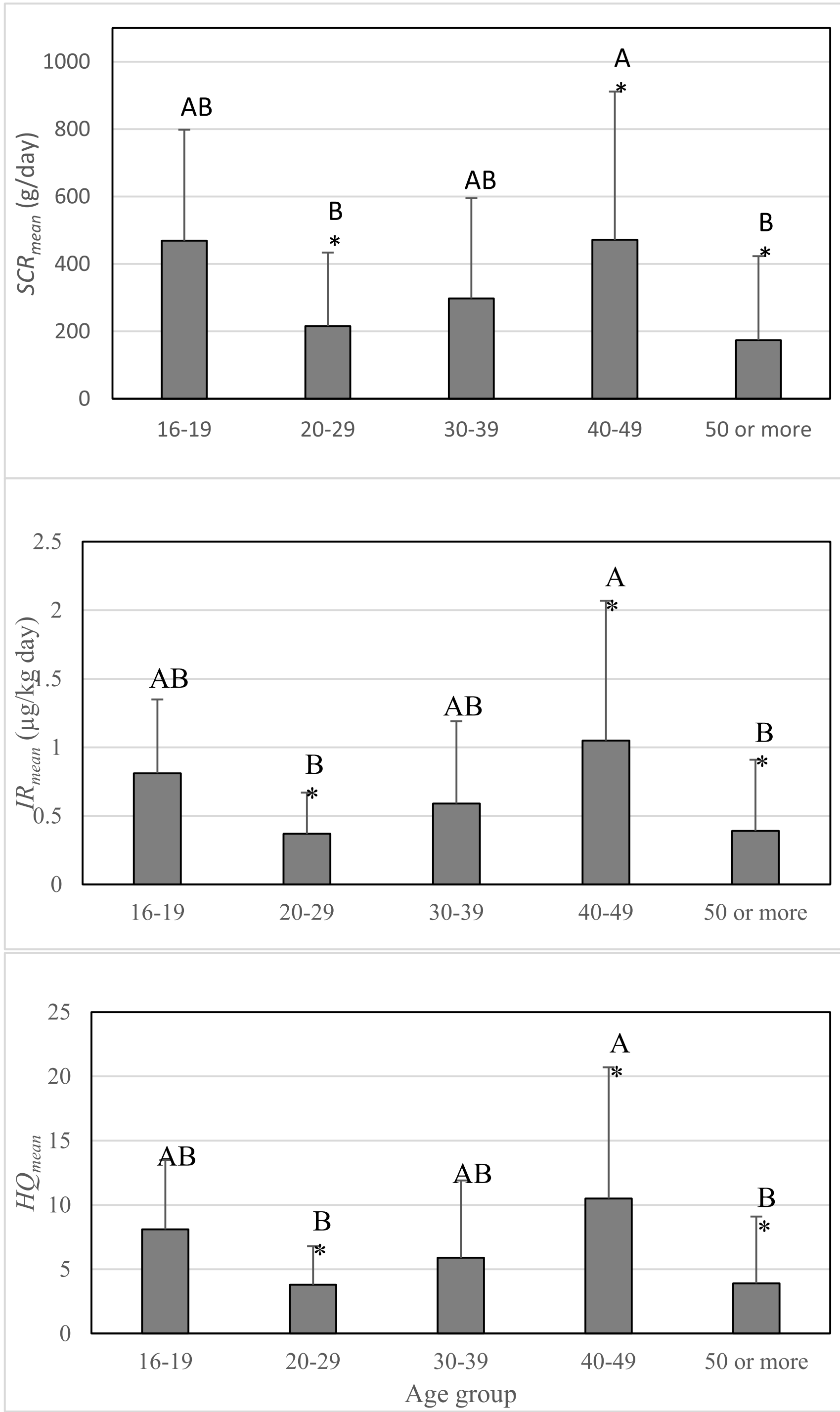

Fig. 2. Differences in SCR_{mean}, IR_{mean} and HQ_{mean} values, grouped by age of interviewed women.

Mazatlan, a coastal city of central Sinaloa (Zamora-Arellano et al., 2017). Mean fish consumption rates in Sonora and Sinaloa were higher than consumption by subsistence population (women) of the U.S. (39.1 g day^{-1}) (Burger, 2002). And higher than fish consumption by females (18–49 years old) from coastal rural communities of Malaysia (136.4 g day^{-1}), the highest fish consumer country in Southeast Asia (Jeevanaraj et al., 2018).

From the total 184 listed preparations of seafood, females selected up to 68 with a mean of 14 ± 8. In general, 58% of interviewed women preferred swimming crabs, followed by mullets (45%), weakfish (45%), blue shrimp cooked (44%) and other 30 seafood products; the least preferred seafood were Gulf grouper (2%), pompano (5%) and Pacific pomfret (6%) (Fig. 3). There were differences in the types of seafood preferred by each age group. Females aged 16–19, preferred breaded blue shrimp, females 20–29 preferred cooked swimming crab, steamed blue shrimp and raw pen shell, the 30–49 group preferred cooked swimming crab, stewed weakfish and steamed blue shrimp, and the 50 or more group preferred cooked swimming crab, stewed mullet and steamed blue shrimp. Also, the rate between raw and cooked fish and shellfish products was higher for young women (0.33 and 0.25 for women 20–29 and 16–19 respectively) and decreased for females 30–39 (0.19), 40–49 (0.11) and older than 50 (0.11).

A simple linear regression was calculated to predict methyl mercury concentrations based on seafood consumption rate SCR_{total}. A weak

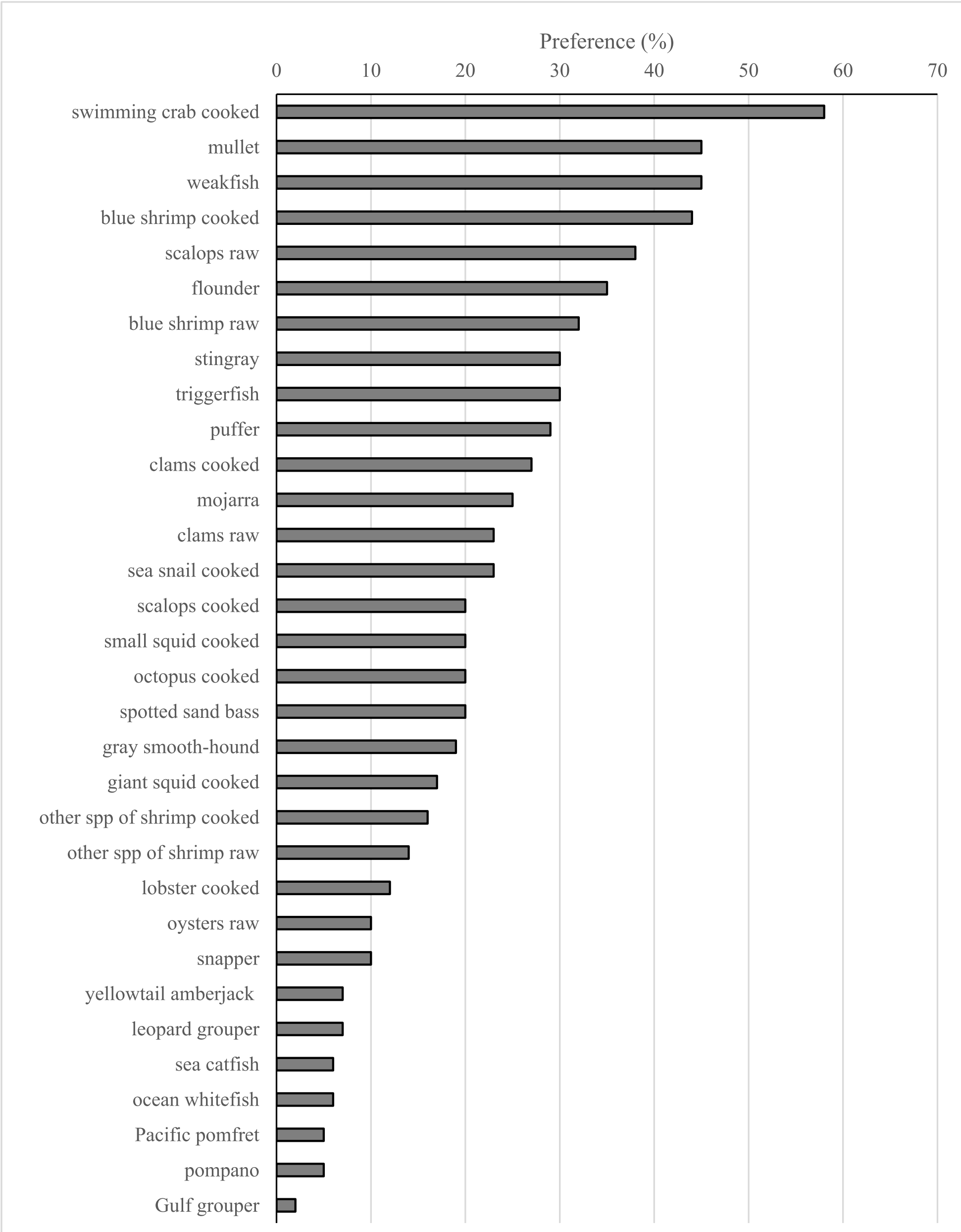

Fig. 3. Seafood preferences among females from the fishing communities of coastal Sonora, Mexico.

correlation was found ($F_{1,107} = 5.21$, $p = 0.02$), with an R^2 of 0.04. Methyl mercury predicted concentration was equal to $2.08 + 0.001(SCR_{total})$ μg g^{-1} (Fig. 4). Results indicate that women who consumed sand bass, yellowtail jack and whiptail stingrays, resulted in higher methyl mercury levels even if consumption was low, and those who consumed other fish like mullets, triggerfish, clams and crabs, had less methyl mercury concentrations even if consumption was high. The lack of correlation between fish consumption and mercury concentrations, was probably due to the high variety of fish and shellfish that the surveyed population consumed. Opposite to the results found in California by Shilling et al. (2010), where freshwater female anglers from the Central Valley who had a mean consumption rate of 38 g day^{-1} and males of 26.4 g day^{-1}, presented a good correlation between fish consumption and mercury intake, this was attributed to the similar fish species consumed.

According to the World Health Organization (FAO/WHO, 2011), consumption of fish provides energy, protein and a range of other important nutrients, including long-chain n-3 polyunsaturated fatty acids (LCn3PUFAs), also eating fish is part of the cultural traditions of many peoples and in some population, fish is a major source of food and essential nutrients. The FAO/WHO report (2011), also states that when comparing the benefits of the LCn3PUFAs with the risks of methylmercury among women of childbearing age, maternal fish consumption lowers the risk of suboptimal neurodevelopment in their offspring compared to women not eating fish. However, for consumption of fish at 100 g day^{-1}, the negative effect of mercury was higher than the positive effects of docosahexaenoic acid (DHA) for any fish that contained more than 0.5 μg g^{-1} of methylmercury. In the present study, consumption was higher than the 100 g day^{-1}, and although mercury concentrations in individual fish and shellfish were lower than 0.5 μg g^{-1}, due to the consumption of several kinds of seafood, the individual amount of methyl mercury ingested was almost 5 times this limit, for which the negative effects of MeHg could exceed the positive effects of seafood. The FDA (2014) recommends a portion of 340 g or 12 ounces per week (48 g day^{-1}) of a variety of fish to obtain the largest development benefits of the fetus. At the coastal communities of

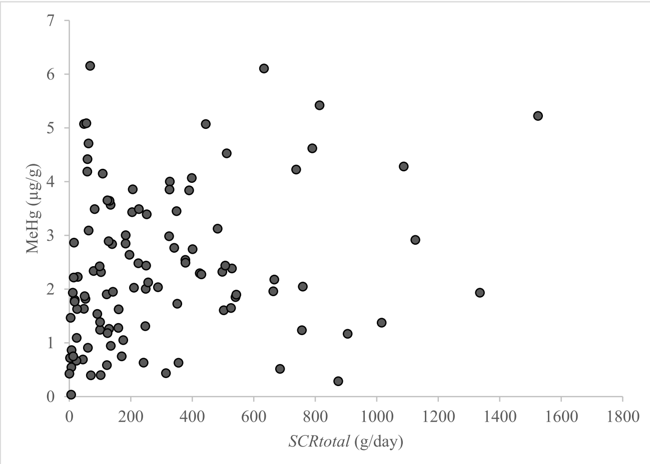

Fig. 4. Correlation between total seafood consumption rate (SCR_{total}) and estimated methyl mercury concentrations in seafood.

Sonora, only 5% of women consumed the recommended amount of fish (between 45 and 55 g day^{-1}), 14% consumed less and the majority (80%) consumed more.

It is fortunate that piscivorous fish like the Gulf grouper, pompano, and yellowtail jack, are not commonly consumed by women at these coastal communities (< 10% of the population), since these species have high mercury concentrations (Table 1). In Sinaloa, the main products consumed by the fishing-related population were shrimp, tilapia, pacific sierra and canned tuna (Zamora-Arellano et al., 2017). Differences between Sonora and Sinaloa food preferences, are due to species availability at each location and conditions, however at both sites, affordability was one of the most important variables to select the product.

3.3. Risk estimation

Methyl mercury ingestion rate (IR_{mean}) varied from 0.002 up to 3.19 µg kg^{-1} day, with a mean of 0.62 ± 0.68 µg kg^{-1} day, 96% of the population had ingestion rates higher than the recommended value of 0.1 µg kg^{-1} day (U.S.EPA, 2001). There was a significant difference in IR_{total} between female age groups (oneway ANOVA, *p-value* = 0.003), with the 40–49 years old group (1.05 ± 1.02) significantly higher compared with the 20–29 group (0.37 ± 0.12) and the 50 or more group (0.39 ± 0.52) (Tukey-Kramer HSD, alpha = 0.05) (Fig. 2). Consequently, the hazard quotient (HQ_{mean}) resulted in values from 0.02 up to 32, with a mean of 6.2 ± 6.8. The majority (83%) of women had HQ_{total} > 1 and presented the same statistical differences as IR_{mean} between the age groups (Fig. 2). The age group with the highest risk was the 40–49 years old population (HQ_{mean} = 10.6), followed by the 16–19 (HQ_{mean} = 8.1), the 30–39 (HQ_{mean} = 5.9), the 50 or more (HQ_{mean} = 3.9) and the lowest risk was presented by the 20–29 (HQ_{mean} = 3.7). It is important to note that methyl mercury in blue crabs composed 93–100% of THg in tissues (Adams and Engel, 2014; Ghaeni et al., 2015) not 50% as was previously considered in the methods section. If this factor is adjusted from the calculations, the HQ_{mean} would increase by 0.2 resulting in a value of 6.4.

Due to the high variability in seafood consumption rates, and consequently high variability in *IR* and *HQ* indices, percentiles of the population are useful to better understand the relationships between consumption and risk (Table 3). Considering a median seafood consumption (50th percentile SCR_{total}); a hazard quotients (HQ_{total}) higher than 1 were reached at the 25th percentile in females 16–19 years old, at the 50th percentile in females 20–29, at the 25th percentile in females 30–39, at the 50th percentile in females 40–49 years old and at the 75th percentile in females older than 50. Therefore, more females 16–19 and 30–39 were at risk compared with females 20–29 and 40–49 and the least risk was found in females older than 50 years old.

Table 3
Total seafood consumption SCR_{total} (g day^{-1}) and total hazard risk (HQ_{total}) from fish and shellfish consumption of different age women from the coast of Sonora, Mexico.

SCR_{total} (g day^{-1})[a] and percentile	Percentile risk (HQ_{total})						
	females 16–19 years old (n = 10)						
	10th	25th	50th	75th	90th	95th	99th
138 (25th)	0.55	0.55	1.26	1.96	1.96	1.96	1.96
498 (median)	0.55	0.9	2.85	3.76	3.76	3.76	3.76
665 (75th)	0.55	1.96	3.76	12.14	12.5	12.15	12.5
790 (90th)	0.55	2.85	7.38	12.32	16.98	16.98	16.98
790 (95th)	0.55	2.85	7.38	12.32	16.98	16.98	16.98
790 (99th)	0.55	2.85	7.38	12.32	16.98	16.98	16.98
	females 20–29 years old (n = 26)						
63 (25th)	0.07	0.17	0.58	1.36	1.37	1.37	1.37
133 (median)	0.11	0.3	1.36	2.99	3.38	3.34	3.34
334 (75th)	0.17	0.73	2.59	3.41	5.59	7.5	7.5
569 (90th)	0.17	0.89	2.81	3.79	5.47	7.5	7.5
810 (95th)	0.18	1.28	3.16	5.09	8.34	9.2	11.02
905 (99th)	0.18	1.32	3.25	6.09	8.82	10.51	11.02
	females 30–39 years old (n = 33)						
100 (25th)	0.03	0.51	1.01	1.91	2.26	2.26	2.26
226 (median)	0.32	0.93	2.18	3.09	4.26	4.55	4.55
393 (75th)	0.52	1.25	2.92	4.12	7.25	7.7	9.01
528 (90th)	0.57	1.81	3.34	5.9	10.32	10.92	13.33
758 (95th)	0.65	2.13	3.68	8.16	11.65	13.36	15.26
760 (99th)	0.68	2.16	3.8	3.88	12.83	15.26	15.26
	females 40–49 years old (n = 22)						
108 (25th)	0.08	0.12	0.36	1.97	2.41	2.41	2.41
389 (median)	0.1	0.36	2.57	4.04	12.02	13.51	13.51
757 (75th)	0.15	1.97	4.03	9.99	14.41	14.52	14.56
1275 (90th)	0.18	2.45	6.63	14.16	28.76	29.25	31.92
1459 (95th)	0.2	2.49	7.17	14.46	29.68	29.94	31.92
1481 (99th)	0.22	2.53	8.05	14.76	29.39	31.66	31.92
	females 50 and more years old (n = 19)						
17 (25th)	0.02	0.05	0.17	0.27	0.3	0.3	0.3
77 (median)	0.02	0.17	0.31	1.25	2.3	2.3	2.3
200 (75th)	0.08	0.27	1.25	2.06	3.15	3.16	3.16
354 (90th)	0.12	0.31	1.93	3.15	8.31	12.37	14.18
378 (95th)	0.13	0.31	1.94	3.51	12.37	14.18	14.18
378 (99th)	0.13	0.31	1.94	3.51	12.37	14.18	14.18

[a] Excluding three outliers: 16–19 (1016 g day^{-1}), 30–39 (1524 g day^{-1}), 50 or more (1088 g day^{-1}).

A high mean hazardous risk (HQ_{mean} = 6.2) in the population surveyed was not expected, since mercury concentrations in fish and shellfish were in general low for this region. However, the high consumption of different seafood products resulted in high methyl mercury ingestion rates and high hazard quotients. Other studies have found similar results in different human populations (MacIntosh et al., 1997; Burger, 2002; Morrissette et al., 2004; Webb et al., 2004; Burger et al., 2005; Bjömberg et al., 2005; Copat et al. 2013, 2014; Ceccato et al., 2016).

Comparing this study with the Sinaloa results (Zamora-Arellano et al., 2017), authors reported high HQ in children from the general population (mean HQ = 10.9), who consumed mainly canned tuna, pacific sierra, shrimp and smoked marlin, and due to their low body mass and high mercury concentrations in these particular species, the risk was higher. On the other hand, females from the fishing related population who consumed shrimp, tilapia, Pacific sierra and canned tuna presented a lower HQ (mean = 1.89), which was lower than the values found for females of the coastal communities of Sonora.

The mean HQ_{mean} found for coastal communities of Sonora corresponded to the 90th percentile HQ for typical female subsistence fish consumer from the U.S.A. who consumed a mean of 39 g day^{-1} of fish (U.S.EPA, 2011). Instead, risks found at the coastal communities of Sonora, were comparable to the population of low income black female subsistence fish consumer in the southeast of the U.S.A., who consumed a mean of 171 g day^{-1} of fish and presented a mean hazard risk (HQ) of 9.4 and up to 56.4. Hazard quotient determined for Malaysian females with high fish consumption was 4.3 ± 1.2 for coastal rural population, and 2.2 ± 0.8 for urban population. Fish that contributed to higher Hg daily dose were silver pomfret, flathead gray mullet, silver whiting, torpedo scad, indian mackerel, pale-edged stingray, fourfinger threadfin and sin croaker (Jeevanaraj et al., 2018). Some of these species were common to the coastal communities of Sonora, Mexico, like the stingray, which also contributed with higher dietary methyl mercury concentrations, and mean hazard quotients were similar between these two distant human populations.

Other studies have shown a lower HQ (< 1) for a variety of commercial fish species from Sinaloa (Ruelas-Inzunza et al., 2011a) and the coast of Guerrero (eastern Pacific) (Spanopoulos-Zarco et al., 2014), considering an average fish consumption of 25 g day^{-1}. However, as mentioned by Ruelas-Inzuna et al. (2011a), higher consumption by fishermen, anglers and their families is of concern because it could result in higher HQs. This hypothesis was confirmed with results from Zamora-Arellano et al. (2017) and the present study.

In Mexico, the majority of women of reproductive age (31.5%) are between 20 and 29 years old (Consejo Nacional de Población, 2011). It was fortunate that fish consumption in this age group was significantly lower than in the rest of the groups (Fig. 2, Table 3) and thus the risks were also lower, although present. There is no clear explanation for the decrease in fish and shellfish consumption in this age group, but it is possible that there is an awareness of the risks of eating fish during pregnancy, however, this hypothesis needs to be tested with more specific interviews.

4. Conclusions

Mean concentrations of mercury in fish and shellfish from the central Gulf of California were in general lower than the MPL, however due to high frequency seafood consumption by women from fishing villages of the coast of Sonora, the risk of methyl mercury exposure was high. The age group that presented the highest risk were women of 40–49 years old, and the lowest risk were for women of 20–29 years old. There were two factors that mitigated the risks of methyl mercury exposure: 1) that the most consumed seafood species had low THg concentrations and 2) that women of reproductive age consumed significantly less seafood than the rest of the age groups. However, in order to lower the risks of methyl mercury exposure, it is recommended to limit seafood consumption in pregnant women to 4 portions of fish per week, preferably of weakfish, tilefish or whitefish, due to their low THg concentrations.

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Acronyms

DL	detection limit
MPL	maximum permissible level (0.5 μg g^{-1})
MeHg	methyl mercury
THg	total mercury
Shellfish	bivalves, mollusks, shrimps, crabs and cephalopods
Seafood	shellfish and fish
TL	Total length
W	Weight
FFQ	Food frequency questionnaire
MCF	Mercury conversion factor
FPCAF	Preparation/cooking adjusting factor
SCR	Seafood consumption rate
SCRtotal	Total seafood consumption per person
SCRmean	Average of total individual seafood consumption
IRtotal	Total daily methyl mercury intake per person
IRmean	Average of total daily mercury intake
HQtotal	Total hazard risk per person
HQmean	Average of total hazard risk
Rfd	Reference dose

Transparency document

Transparency document related to this article can be found online at https://doi.org/10.1016/j.fct.2018.07.029.

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