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## Linking fish consumption patterns and health risk assessment of mercury exposure in a coastal community of NW Mexico

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### ABSTRACT

We aimed at characterizing the methylmercury (MeHg) exposure through fish consumption in two populations (common general and fishing-related population (FRP)) using a probabilistic health risk assessment in children, women of childbearing age, and adults in Mazatlán. The hazard quotients (HQs) were obtained from fish consumption, defined through a survey, and the levels of mercury in fishery products, obtained from published information. The average fish ingestion rate ( $IR_{food}$ ) was higher in the FRP ( $167.85 \text{ g d}^{-1}$ ) than in the general population (GP) ( $140.9 \text{ g d}^{-1}$ ). However, HQs were significantly ( $p < 0.05$ ) higher in the GP (ranging from 0.18 to 10.91) compared to the FRP (0.20 to 2.48); significant differences were also found among groups of both populations. Remarkably, children in both populations exhibited the highest proportions of risk, reaching up to 97% in GP. For all populations, fish consumption was the most important variable influencing MeHg exposure. Overall, for MeHg exposure, there is no safe level of fish consumption without risk, and actions should be taken to mitigate possible risk; further research with current data is needed to assess potential health risks associated with MeHg exposure, particularly in children.

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fishery products; mercury exposure; Gulf of California

## Introduction

Several fish-eating communities around the world have been reported with high levels of Hg exposure. Some of these are located near the coasts, major lakes, and rivers, where fish consumption is an integral part of their diet (Chan 2011). However, some factors can influence Hg exposure, such as the species of fish consumed (*i.e.*, species with a higher trophic level tend to accumulate more Hg than species of lower trophic levels), rate of consumption, the presence of a pollution source, and the geochemical characteristics of the water bodies. Mercury exposure due to fish consumption is likely to be in its methylated form, as it has been reported that up to 90% of Hg in fish muscle is present as methylmercury (MeHg) (Freije and Awadh 2009). MeHg is a

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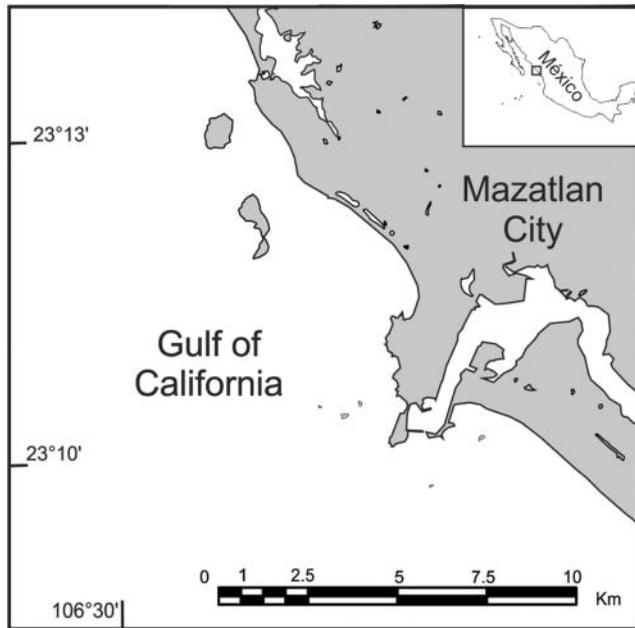
neurotoxic compound; therefore, the central nervous system is the principal target, especially in fetus (Díez 2009), infants (1–24 months), and children (2–12 years old) (Driscoll *et al.* 2013). One of the most important and best studied end points of MeHg toxicity is the neurodevelopmental effect in the children of woman who have consumed fish and seafood during pregnancy (NAS 2000). Usually, maternal hair is used as a bioindicator of exposure, and then the effects on exposed children during pregnancy or nursing have been associated to neurological impairment that decreases learning capacity and motor coordination. A concentration of 6 ppm of MeHg in maternal hair has been established as a threshold of neuroconductual dysfunction in children; at levels of 10–20 ppm, a decrease in memory and motor function occurs; above 20 ppm, there is a delay in psychomotor activity (Grandjean *et al.* 1995). Consequently, fish consumption by pregnant women, children, and women of childbearing age is of particular concern due to potential consequences for present and future generations. In order to protect the fetus and development, the USEPA/FDA (2014) recommends a consumption of 8–12 oz of fish per week to children and pregnant women. However, in our country, there are no such recommendations or advisories to protect the most vulnerable sectors of the population from the effects of mercury. The scarce information on mercury in fishery products makes it difficult to establish safe levels of consumption.

In contrast, fish consumption is desirable as it provides an excellent source of high-quality proteins and fatty acids such as omega-3. The intake of fish has been related to a decreasing risk of coronary heart disease and stroke, and as a promoter of growth and development (FAO/WHO 2010). In fact, worldwide fish consumption increased from 9.9 kg per capita in 1960 to 19.2 kg per capita in 2012 (FAO 2014). In Mexico, marine fisheries are an important economic activity; therefore, higher fish consumption in coastal communities compared to inland communities or the national average consumption ( $32 \text{ g}^{-1} \text{ d}^{-1}$ ; SAGARPA 2013) can be expected. Also, fish consumption is expected to be higher in fishing-related populations (FRPs) compared with common population, although consumption patterns may be different due to accessibility and socioeconomic factors. In this context, to assess MeHg risk related to fish consumption, the objectives of the present study were (1) to assess consumption patterns of fishery products in a typical coastal community of NW Mexico (Mazatlán, state of Sinaloa), (2) to carry out a risk assessment based on existing Hg levels of fishery products for human consumption, and (3) to identify the main factors related to Hg intake in the different groups of interest through a sensitivity analysis. A probabilistic risk approach was followed to estimate the portion of population likely to be at risk due to MeHg exposure.

## Methods and materials

### Study area

The Sinaloa state is located in NW Mexico ( $27^{\circ}02' - 22^{\circ}29' \text{ N}$  and  $105^{\circ}23' - 109^{\circ}28' \text{ W}$ ) and has a coastal line of 640.17 km (Figure 1). Mazatlán is the main coastal city, with its main activities being related to tourism, fisheries, and industry; its harbor has the second largest fishing fleet in Mexico and the biggest tuna processing industry in Latin America. Besides, there are shipyards and other fishing-related industries. According to SAGARPA (2013), there are 546 open sea fishing vessels (469 are used for shrimp trawling, 30 for tuna, 9 for sardines, and the rest for other resources) in Sinaloa, while the artisanal fisheries account for 11,198 boats (around 70% of large vessels and fishing boats are located in the municipality of Mazatlán). The population related to fishing activities accounts for 273,000 with 17% being fishermen (SAGARPA 2013).



**Figure 1.** Localization of the surveyed area in the state of Sinaloa, Mexico.

The main anthropogenic sources of trace metals in Mazatlán are domestic and industrial wastes, landfills (receiving around 450 ton per day; INEGI 2013), and mining, mainly extracting lead, iron, zinc, silver, gold, and copper (SE/SGM 2014). Another contribution of Hg and other metals is the atmospheric transport from near and remote areas (Pirrone *et al.* 2010). Additionally, the area has experienced a pronounced Hg enrichment due to energy generation. From dating of sediment cores, it has been established that since 1968, there has been Hg enrichment and the most probable source is atmospheric deposition. A thermoelectrical power plant in Mazatlán started operations in 1966 and it is likely that Hg accumulation in sediments of the region originated due to the atmospheric discharges of this power plant (Ruiz-Fernández *et al.* 2009). With respect to natural sources, the geology of the area indicates that Hg is not enriched (Rytuba 2003).

### **Food frequency survey**

The food frequency survey is an inexpensive, fast, and easy method that allows obtaining information of long-term consumption habits (Cade *et al.* 2002). It requires a lesser effort from the interviewed than other methods; it does not alter the pattern of habitual consumption and allows extracting information on the influence of seasonal variability. These surveys are useful to classify individuals of a population according to their consumption, which allows comparisons and identification of high-risk behaviors (Trinidad-Rodriguez *et al.* 2008). Nevertheless, some limitations of the food surveys need to be considered, such as the recall bias due to the influence of the current diet, and the uncertainties associated with the estimation of portion size and the identification of the species consumed. The survey could also fail to identify sporadic events of consumption of fishery products with high Hg content. In the present study, fish and shellfish consumption information was obtained through a food frequency

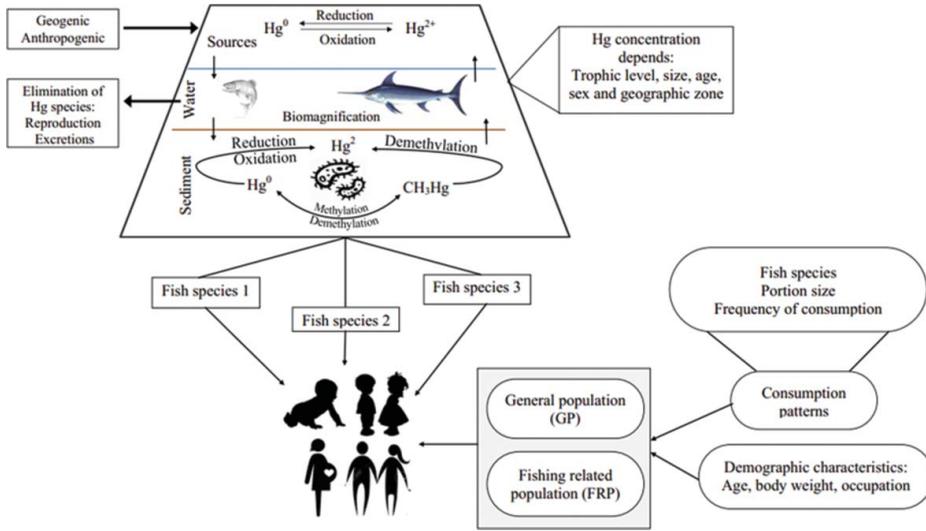
survey applied in Mazatlán, Sinaloa, in the summer of 2014. The food frequency questionnaires were carried out following the procedure described in García-Hernández *et al.* (2013), based on the food composition tables developed by Ortega *et al.* (1999). Adult males and females were invited to participate in the survey by direct contact in the different fish markets of the city; they were also asked to authorize their participation and of their children and to provide their addresses. The questionnaire was applied directly by reading the questions and annotating the answers. The respondents agreed to provide personal information as well as sociodemographical and consumption practices. To characterize the consumption patterns of fishery products, we considered 48 food types; the frequency of consumption was recorded as daily, weekly, monthly, annually, or rarely. Portion sizes were standardized according to a food dictionary compiled at CIAD (Ortega *et al.* 1999) and were as follows: 80 g for fish fillet (the average weight of a fish fillet in a dish), 100 g for shrimp, 12 oysters, and 130 g for canned tuna (it corresponds to the drained mass of a can). Information such as age and last school grade was included. Body weight was obtained directly from the surveyed people; in the case of children, the weight data corresponded to the last record of health cards provided by the governmental health system. Additionally, people were asked about their awareness on the toxicity of mercury and its presence in fishery products (Supplementary Material, Table S1). In order to be representative of the population of Mazatlán, the number of questionnaires ( $N$ ) is estimated according to Eq. (1) (Henry, 1990):

$$N = \frac{z^2 pq}{e^2} \quad (1)$$

where  $z$  is the confidence level ( $95\% = 1.96$ ),  $pq$  is the population variance (0.25), and  $e$  is the sampling error (0.05). Although a total of 384 questionnaires were applied, only 370 were considered valid for this study (14 questionnaires were rejected because of the lack of information related to weight or age); the population surveyed was divided into two groups in response to their main economic activity. Group 1 consisted of FRP ( $n = 161$ ), defined as the population of either men, women, or children who are related to some aspect of a fishery activity, either as an independent fishermen or as part of an industrial fishing ship, and their families. Individuals from this group were separated as “children A” (3–10 years old,  $n = 9$ ), “children B” (11–15 years old,  $n = 13$ ), “women C” (women in childbearing age, 16–40 years old,  $n = 45$ ), and “rest of population D” (men > 16 years old,  $n = 86$ ; women > 41 years old,  $n = 8$ ). Group 2 included general population (GP,  $n = 209$ ) defined as the rest of the population who does not have any connection with the fishing activities. Individuals from this group were also separated as “children A” (3–10 years old,  $n = 11$ ), “children B” (11–15 years old,  $n = 26$ ), “women C” (women of childbearing age, 16–40 years old,  $n = 55$ ) and “rest of population D” (men > 16 years old,  $n = 81$ ; women > 41 years old,  $n = 36$ ).

### **Mercury data and risk assessment**

The risk assessment conceptual model is described in Figure 2. Only the fishery products with Hg data in edible portion from the published literature available were considered (Table 1). When more than one data per product were available, average data were used. A model for non-carcinogens (HQ) was considered according to USEPA (1997a). The individual food ingestion rate ( $IR_{food}$ ; expressed as  $g\ d^{-1}$ ) is determined (Eq. (2)), considering



**Figure 2.** Conceptual model of mercury exposure through fish consumption.

the preferences of consumption for each person interviewed.

$$IR_{food} = \sum \frac{\text{portion size (g)} \times \text{number of portions} \times \text{frequency of consumption}}{365} \quad (2)$$

Additionally, according to the criterion by WHO (2008), groups with intakes ( $IR_{food}$ ) over 100 g of fish and shellfish per day are considered as the threshold between high and low consumptions, especially for women of childbearing age because of the risk to the developing fetus.

To determine the individual food ingestion dose ( $Dose_{food\ ingestion}$ ), a probability distribution was used, rather than a single value for the variables involved, using the Oracle Crystal Ball 11.1.2.3.500 software and a Monte Carlo simulation with 10,000 iterations (USEPA 2001). Furthermore, because of the diversification and independent variation of the involved parameters, the distribution was defined as a probability density function; such a function is derived from a limited number of observations. This approach allows a sufficient number of samples for modeling with an elevated certainty due to the generation of random numbers for each parameter (USEPA 1997b). The food ingestion dose is calculated according to Eq. (3):

$$Dose_{food\ ingestion} = \sum \frac{C_{Food} \times IR_{Food} \times AF_{GIT} \times D_{Days}}{BW \times 365} \quad (3)$$

where  $C_{Food}$  refers to the concentration of the contaminant in food ( $\text{mg kg}^{-1}$  wet weight); a conversion of Hg concentrations from dry weight ( $Hg_{dw}$ ) to fresh weight ( $Hg_{fw}$ ) is done according to Eq. (4) (Magalhães *et al.* 2007):

$$Hg_{fw} = Hg_{dw} * (100 - \% \text{ water content} / 100) \quad (4)$$

**Table 1.** Mercury concentrations ( $\mu\text{g g}^{-1}$  dry weight) reported in edible tissue of fishery products from NW Mexico.

Scientific name	Common name	Hg	N	Reference
Crustaceans				
<i>Litopenaeus vannamei</i>	Shrimp <sup>1</sup>	0.31 $\pm$ 0.21	13 <sup>a</sup>	Delgado-Alvarez <i>et al.</i> (2015)
Elasmobranches				
<i>Gymnura marmorata</i>	California butterfly ray <sup>2</sup>	0.71	1	Ruelas-Inzunza <i>et al.</i> (2013)
<i>Carcharhinus leucas</i>	Bull shark <sup>3</sup>	0.20 $\pm$ 0.09	1	Ruelas-Inzunza and Páez-Osuna (2005)
<i>Sphyrna lewini</i>	Scalloped hammerhead <sup>3</sup>	4.84 $\pm$ 0.05	1	Ruelas-Inzunza and Páez-Osuna (2005)
Molluscs				
<i>Crassostrea corteziensis</i>	Cortez oyster <sup>4</sup>	0.06 $\pm$ 0.02	24 <sup>a</sup>	Jara-Marini <i>et al.</i> (2010)
<i>Crassostrea gigas</i>	Pacific oyster <sup>4</sup>	0.06 $\pm$ 0.91	6	Osuna-Martínez <i>et al.</i> (2010)
Fishes				
<i>Mugil cephalus</i>	Gray mullet <sup>5</sup>	0.06 $\pm$ 0.03	15	Reimer and Reimer (1975)
<i>Lutjanus colorado</i>	Colorado snapper <sup>6</sup>	0.53 $\pm$ 0.27	10 <sup>a</sup>	Ruelas-Inzunza <i>et al.</i> (2008)
<i>Diapterus peruvianus</i>	Peruvian mojarra <sup>7</sup>	0.58 $\pm$ 0.18	5 <sup>a</sup>	Ruelas-Inzunza <i>et al.</i> (2008)
<i>Scomberomorus sierra</i>	Pacific sierra <sup>8</sup>	0.64 $\pm$ 0.06	1 <sup>a</sup>	Ruelas-Inzunza <i>et al.</i> (2008)
<i>Trachinotus paitensis</i>	Paloma pompano <sup>9</sup>	1.42 $\pm$ 0.26	3 <sup>a</sup>	Ruelas-Inzunza <i>et al.</i> (2008)
<i>Gerres cinereus</i>	Yellowfin mojarra <sup>7</sup>	0.82 $\pm$ 0.16	6 <sup>a</sup>	Ruelas-Inzunza <i>et al.</i> (2008)
<i>Mugil curema</i>	White mullet <sup>5</sup>	0.47 $\pm$ 0.69	11 <sup>a</sup>	Ruelas-Inzunza <i>et al.</i> (2008)
<i>Mugil cephalus</i>	Striped mullet <sup>5</sup>	0.07 $\pm$ 0.05	7 <sup>a</sup>	Ruelas-Inzunza <i>et al.</i> (2008)
<i>Paralichthys woolmani</i>	Speckled flounder <sup>10</sup>	0.68 $\pm$ 0.29	3 <sup>a</sup>	Ruelas-Inzunza <i>et al.</i> (2008)
<i>Caranx caninus</i>	Pacific crevette Jack <sup>11</sup>	3.32 $\pm$ 3.15	2 <sup>a</sup>	Ruelas-Inzunza <i>et al.</i> (2008)
<i>Sphoeroides annulatus</i>	Bullseye puffer <sup>12</sup>	0.98 $\pm$ 0.49	15 <sup>a</sup>	Ruelas-Inzunza <i>et al.</i> (2008)
<i>Pomadasyd branickii</i>	Sand grunt <sup>13</sup>	1.17 $\pm$ 0.06	2 <sup>a</sup>	Ruelas-Inzunza <i>et al.</i> (2008)
<i>Thunnus albacares</i>	Yellowfin tuna (canned in oil) <sup>14</sup>	0.26 $\pm$ 0.12	42	Ruelas-Inzunza <i>et al.</i> (2011b)
<i>Thunnus albacares</i>	Yellowfin tuna (canned in water) <sup>14</sup>	0.36 $\pm$ 0.19	46	Ruelas-Inzunza <i>et al.</i> (2011b)
<i>Tetrapturus audax</i>	Striped marlin <sup>15</sup>	5.63 $\pm$ 0.60	13	Soto-Jiménez <i>et al.</i> (2010)
<i>Larimus argenteus</i>	Silver drum <sup>16</sup>	0.90 $\pm$ 0.75	67	Ruelas-Inzunza <i>et al.</i> (2012)
<i>Selar crumenophthalmus</i>	Bigeye scad	0.55 $\pm$ 0.49	3	Ruelas-Inzunza <i>et al.</i> (2012)
<i>Trachinotus kennedyi</i>	Brackblotch pompano <sup>9</sup>	0.11 $\pm$ 0.08	34	Ruelas-Inzunza <i>et al.</i> (2012)
<i>Diapterus peruvianus</i>	Peruvian mojarra <sup>7</sup>	2.55 $\pm$ 1.17	123	Ruelas-Inzunza <i>et al.</i> (2012)
<i>Cyclopsetta querna</i>	Toothed flounder	0.39 $\pm$ 0.23	10	Ruelas-Inzunza <i>et al.</i> (2012)
<i>Ophioscion strabo</i>	Squint-eye croaker	1.65 $\pm$ 1.17	2	Ruelas-Inzunza <i>et al.</i> (2012)
<i>Diplectrum pacificum</i>	Inshore sand perch <sup>17</sup>	0.44 $\pm$ 0.35	2	Ruelas-Inzunza <i>et al.</i> (2012)
<i>Isopisthus remifer</i>	Silver weakfish <sup>18</sup>	1.77 $\pm$ 0.63	12	Ruelas-Inzunza <i>et al.</i> (2012)

<sup>a</sup>Pooled samples. Average Hg value, 0.23  $\mu\text{g g}^{-1}$  (using the probabilistic approach).

<sup>1-18</sup>Seafood items used for risk assessment.

We used an average of 70% for water content (Ruelas-Inzunza *et al.* 2011a);  $IR_{food}$  is the food ingestion ( $\text{g d}^{-1}$ ) rate;  $AF_{GIT}$  is the absorption factor for the gastrointestinal tract; a value of 1 was used for a preliminary risk assessment as recommended by Health Canada (2004);  $D_{Days}$  is the number of days in a year that food from the site is ingested (1–365);  $BW$  is the body weight of the consumer. The  $Dose_{food\ ingestion}$  is expressed as  $\text{mg Hg kg}^{-1}$  of body weight  $\text{d}^{-1}$ .

Finally, the risk was estimated according to the hazard quotient (HQ) using a probabilistic approach (Eq. (5)):

$$HQ = \frac{Dose_{food\ ingestion}}{RfD} \quad (5)$$

where RfD indicates the reference dose, *i.e.*, the concentration of a pollutant expressed on a body weight basis ( $\text{mg kg}^{-1}$  of body weight), which can be ingested over a lifetime without appreciable health risk (WHO 2008). If the calculated HQ is greater than 1, it indicates that a potential risk to human health exists (USEPA 2001). For MeHg, the RfD of 0.0001  $\text{mg kg}^{-1}$  body weight  $\text{d}^{-1}$  was used (USEPA 2000). Details of the calculated parameters are given in

**Table 2.** Significant differences ( $p < 0.05$ ) of HQs between groups and populations were defined by the Kruskal–Wallis non-parametric ANOVA using GraphPad Prism 7.0. Additionally, a tornado chart (optional output of Crystal Ball software) was made in order to identify and rank the most important sources of variability of Hg ingestion (USEPA 2001).

In many probabilistic studies, there are limitations of diverse type; in our study, it is worth mentioning several uncertainties. In the health risk model, the distribution of Hg concentrations in fishery products was a log-normal one, perhaps because of the heterogeneity of samples (crustaceans, molluscs, and fishes). Another limitation of the study is related to the time when the questionnaires were applied, *i.e.*, the survey was carried out in the summer but there were changes in the habits of consumption because of food availability and costs throughout the year. Another key issue is the ratio of MeHg to Hg. It has been concluded that in muscle of fish, the organic form of Hg predominates (>90%); however, this is not the case with crustaceans and cephalopods where MeHg contribution is lower (<60%) than that in fish (Moon *et al.* 2011).

## Results and discussion

### *Population characteristics and perception of Hg in fish*

In the GP group, 46% of surveyed people were males and 54% were females, while in the FRP group, 60% were males and 40% were females. Details of age, education, and occupations of each population are provided in Table 3. The acknowledgment of Hg as an environmental and fish pollutant was (considering women and man) 45 and 36% in the GP group, respectively, contrasting with 32 and 19% in the FRP group. This level of awareness is similar to that reported elsewhere. In a study in Wyoming (USA), only 15% of the survey respondents had the knowledge of Hg as a fish pollutant (Johnston and Snow 2007). A similar study in Japan showed that 50% of women in childbearing age knew that fish can contain contaminants which can have adverse effects; however, 67.6% of women would not change their fish intake even when they know Hg can affect newborns (Chien *et al.* 2010). In another study conducted in 12 states of the United States (Knobeloch *et al.* 2005), the authors focused solely on women, reporting that the knowledge about the warnings about Hg content in fish increases as age increases (7% in women aged 18–25 years, 18% in women aged 26–35 years, and 27% in women aged 36–45 years). The authors indicate that in some cases, age and educational and socioeconomic levels can be a predictor about fish consumption and awareness about the occurrence of other contaminants in fishery products. In our study, no pregnant women were surveyed, but the educational level could have played a role in the Hg awareness, but this is not necessarily related to a lower Hg exposure. For instance, Dong *et al.* (2015) observed an increase in Hg exposure with the education level, and they associated it with the awareness of the health benefits related to fish consumption. In our study, the higher education level in GP may be associated not only with the awareness of health benefits, but also to greater purchasing power for commodity goods such as tuna, smoked marlin, and other high priced fish. In contrast, FRP had higher fish consumption (see later), but with a more diverse fish diet of locally available fish.

### *Consumption patterns*

The average daily consumption for each of the 48 items is presented in Supplementary Table S4. In general, the GP consumed 75.5 kg of fish products annually ( $207 \text{ g d}^{-1}$ ), while



Table 2. Parameters used in the health risk model.

Parameter	Symbol	Units	Distribution	General population		Fishing-related population	
				Low consumption	High consumption	Low consumption	High consumption
Age							
Children A		Years	Normal	7 ± 2	8 ± 4	7 ± 3	—
Children B		Years	Normal	13 ± 2	13 ± 2	13 ± 1	13 ± 1
Women C		Years	Normal	25 ± 8	23 ± 6	28 ± 7	23 ± 7
Rest D		Years	Normal	35 ± 15	38 ± 16	39 ± 14	39 ± 14
Weight							
Children A	BW	Kg	Normal	27 ± 9	42 ± 35	31 ± 13	—
Children B	BW	Kg	Normal	53 ± 10	51 ± 9	47 ± 6	54 ± 9
Women C	BW	Kg	Normal	62 ± 10	59 ± 8	61 ± 13	62 ± 12
Rest D	BW	Kg	Normal	71 ± 15	75 ± 12	79 ± 14	82 ± 12
Consumption							
Children A	IR <sub>food</sub>	g d <sup>-1</sup>	Log normal	36 ± 25	189 ± 61	52 ± 32	—
Children B	IR <sub>food</sub>	g d <sup>-1</sup>	Log normal	53 ± 24	249 ± 146	49 ± 24	247 ± 150
Women C	IR <sub>food</sub>	g d <sup>-1</sup>	Log normal	48 ± 22	203 ± 73	62 ± 25	202 ± 161
Rest D	IR <sub>food</sub>	g d <sup>-1</sup>	Log normal	47 ± 23	303 ± 296	47 ± 27	516 ± 510
Dose food ingestion							
Children A	DI	mg Hg kg <sup>-1</sup> d <sup>-1</sup>	Log normal	0.00006 ± 0.0003	0.00044 ± 0.03585	0.00013 ± 0.00232	—
Children B	DI	mg Hg kg <sup>-1</sup> d <sup>-1</sup>	Log normal	0.00005 ± 0.00009	0.00108 ± 0.0213	0.00003 ± 0.00003	0.00019 ± 0.00019
Women C	DI	mg Hg kg <sup>-1</sup> d <sup>-1</sup>	Log normal	0.00003 ± 0.00003	0.00011 ± 0.00011	0.00003 ± 0.00005	0.00019 ± 0.00025
Rest D	DI	mg Hg kg <sup>-1</sup> d <sup>-1</sup>	Log normal	0.00002 ± 0.00009	0.00023 ± 0.00028	0.00002 ± 0.00002	0.00025 ± 0.00046
Average consumption	IR <sub>food</sub>	g d <sup>-1</sup>	Log normal	140.9			167.85
Absorption factor	A <sub>fGIT</sub>	No unit	Constant		1 <sup>a</sup>		
Hg concentration	C <sub>food</sub>	mg kg <sup>-1</sup>	Log normal		0.34 ± 0.37 <sup>b</sup>		
Days of exposure	D <sub>days</sub>	D	Log normal		1 – 365		
Reference value							
Methylmercury	RfD	mg kg <sup>-1</sup> d <sup>-1</sup>	Constant		0.0001 <sup>c</sup>		

The ages of the groups are as follows: children A (3–10 years old), children B (11–15 years old), women C (women of childbearing age 16–40 years old) and the rest of population D (men >16 years old, and women >41 years old).

<sup>a</sup>According to Health Canada (2004).

<sup>b</sup>Data from Table 1 (μg g<sup>-1</sup> wet weight).

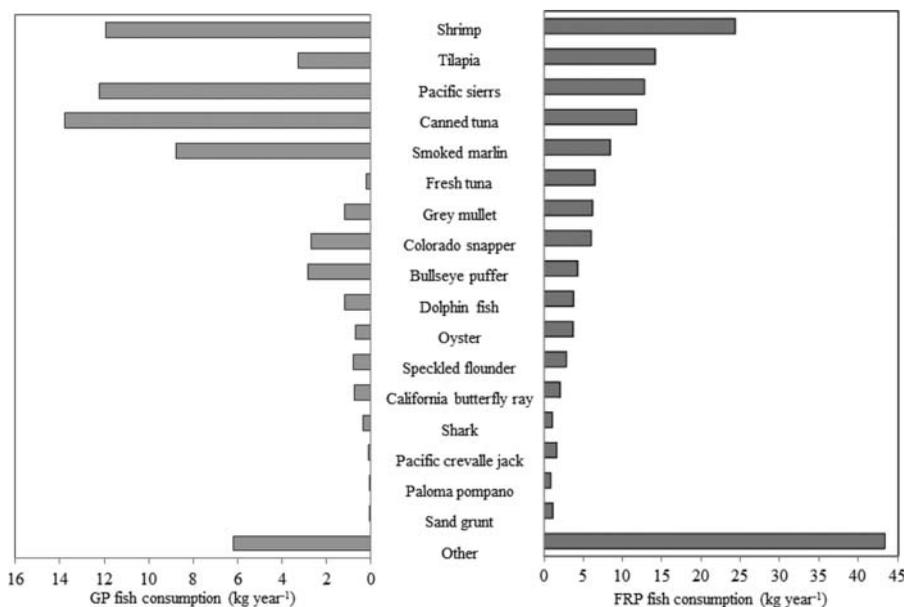
<sup>c</sup>RfD – reference dose by USEPA (2000).

**Table 3.** Demographic composition (percentage) of survey respondents.

Demographic characterization	GP		FRP	
	Male	Female	Male	Female
Age				
3–15	7	11	6	7
16–19	11	12	4	11
20–40	17	14	28	17
>41	11	17	22	5
Total	46	54	60	40
Education				
Primary	4	8	11	4
High school	6	8	19	4
Some college	7	14	14	12
Currently studying	16	11	8	7
Bachelor's degree	10	10	7	12
Graduate studies	2	2	—	—
Not answer	1	1	1	1
Occupation				
Student	30	31	17	19
Employed	12	10	15	14
Housewife	—	12	—	6
Unemployed	0.5	—	—	—
Retired	0.5	0.5	1	—
Fisherman	—	—	27	—
Other	3	0.5	—	1
Knowledge of mercury as a pollutant				
Yes	22	23	12	20
No	24	31	48	20
Knowledge that fishery products can contain mercury				
Yes	18	18	10	9
No	28	36	50	31

GP = General population; FRP = Fishing related population.

the FRP consumed almost the double (152.2 kg of fish product per year or 423 g d<sup>-1</sup>). The fish products mostly consumed by GP were canned tuna, Pacific sierra, shrimp, and smoked marlin. In turn, FRP consumed mostly shrimp, tilapia, Pacific sierra, and canned tuna (Figure 3). The risk was calculated with 18 food items that had available Hg data (shown in Table 2 as  $IR_{food}$ ). In this context, the average  $IR_{food}$  for the GP was 140.9 g d<sup>-1</sup> and for the FRP was 167.85 g d<sup>-1</sup>; these values represent 68 and 39% of the total fish consumption for GP and FRP, respectively (Supplementary Material, Tables S2–S5). Similar to GP in the present study, Ruiz-Guzman *et al.* (2014) reported in a population of Colombia that only four fish species represent 63–72% of the total fish consumption and their consumption rate was 148.33 g d<sup>-1</sup>. As mentioned above, the food items were selected on the basis of their rate of consumption and availability of published information of Hg concentration in the edible portion. On the other hand, in Mexico, the national average consumption of fishery products is 32 g d<sup>-1</sup> (SAGARPA 2013); therefore, even if only 18 items are considered, the consumption in the local population is well above the national average consumption. As can be seen, fish consumption varies in different populations around the world; for example, Dong *et al.* (2015) reported a fish consumption of 58 g d<sup>-1</sup> for anglers in Grand Lake watershed (Oklahoma, USA), but USEPA (2011a) reported a fish consumption of 102 g d<sup>-1</sup>. López-Barrera and Barragán-Gonzalez (2016), in another study in Colombia, reported a consumption of 227 g d<sup>-1</sup> in adults. The Hg values reported in the different studies are above their national average (FAO 2014). Regarding the children in the present study,



**Figure 3.** Annual fish consumption and preference of consumption of general population and fishing-related population.

they showed relatively high fish consumption, ranging from 36 to 249 g d<sup>-1</sup>. These values were also higher than those reported in children from Japan with an average consumption of 52.6 g d<sup>-1</sup> (Zhang *et al.* 2009), and in children (3–10 years old) of an Italian coastal population, with an average consumption of 67 g d<sup>-1</sup> (Brambilla *et al.* 2013).

### MeHg exposure estimation

Fetus and young children are especially susceptible to MeHg exposure; therefore, women in childbearing age and pregnant woman are a group of interest given the potential of transferring the toxicant to the developing baby. Additionally, the segment of the population with high fish and seafood consumption could also become vulnerable as it is likely to be exposed to higher levels of MeHg. The allowable daily intake established for children under 15 years old is 0.1 μg kg<sup>-1</sup> of body weight of MeHg (0.0001 mg kg<sup>-1</sup>) (USEPA 2011b), whereas Health Canada sets a maximum of 0.2 μg kg<sup>-1</sup> (0.0002 mg kg<sup>-1</sup>). In the present study, none of the groups with low consumption exceeded the value of 0.0001 mg kg<sup>-1</sup>, except for “children A” of the FRP, whose values were 30% higher. For the populations with high consumption, all groups were above 0.0001 mg kg<sup>-1</sup>, particularly the groups of GP, where children (A and B) exceeded 4–11 times the established daily dose. This indicates that the exposure to MeHg in children is in fact higher than in adults; therefore, the potential for adverse health effects increases (WHO 2008). Likewise, in 50% of Colombian children, the allowable daily intake was exceeded 29-fold (Ruiz-Guzman *et al.* 2014) and a similar situation has been reported in Marrugo-Negrete *et al.* (2013) where 90% of children were above the tolerable intake. In this regard, FAO/WHO (2010) indicates that “among infants, young children and adolescents, the available data are currently insufficient to derive a quantitative framework of the health risks and health benefits of eating fish.” Nevertheless, our results of

children exposure become a matter of concern due to their developing nervous systems, which makes them susceptible to adverse neurological effects (Grandjean *et al.* 2004).

### **Risk characterization**

In Mexico, the legislation refers to the presence of MeHg in fishery products; for tuna, stripped marlin, grouper, and bonito, the limit is  $1 \mu\text{g g}^{-1}$  wet weight and for the rest of fishery products, the limit is  $0.5 \text{ mg kg}^{-1}$  wet weight (NOM-242-SSA-2009). These limits, however, do not necessarily help to provide dietary advices to reduce MeHg exposure, highlighting the need for risk characterization studies. Existing studies indicate that some Mexican populations may have relatively high levels of fish consumption (Ruelas-Inzunza *et al.* 2011a; Basu *et al.* 2014). However, as the deterministic estimates followed by Ruelas-Inzunza *et al.* (2011a) failed to show risk for most fish species, even when some species were reported with high Hg content, a more detailed probabilistic approach was adopted in the present study. This approach is desirable to reduce uncertainty by using the variability of measurements and the natural fluctuations arising from physiological reasons, *i.e.*, differences in body weight, intake rates, exposure frequency, as well the variability in chemical concentrations (USEPA 2001; Dong *et al.* 2015). Results of the present study indicate significant differences ( $p < 0.01$ ) in risk between the GP and FRP groups, as well as among the subgroups (Table 4). These differences result from the number of iterations used ( $n = 10,000$ ) and consequently the standard error is reduced ( $p > 0.01$ ). Low consumption groups of GP and FRP had HQs  $< 1$ , except for “children A” of FRP (HQ = 1.55). However, the probabilistic approach suggested that only the group “rest of population” D (men  $> 16$  years old and women  $> 41$  years old) with low consumption for GP and FRP showed no risk; for the rest of the groups (low consumption), the percentage of population at risk ranged from 4 to 51%. Interestingly, the higher risks in the low consumption groups were “children A” (13% for GP and 51% for FRP). In contrast, high consumption groups had HQ values above 1, ranging from 1.08 to 10.91 for GP, and from 1.88 to 2.48 for FRP. The percentage of population at risk ranged from 39 to 97%, being “children B” of GP, the group that exhibited the highest percentage (97%; average HQ = 10.91). The percentages of population at risk in the present study are higher than those reported in North America. Similarly, Basu *et al.* (2014) indicated that Hg exposure of pregnant women and children in Mexico City was estimated to be three to five times greater than that in the US and Canada. In a study in Wyoming, the population exceeding the exposure limits was 11.7% for children, 7.9% for women in childbearing age, and 2.2% for the rest of the population (Johnston and Snow 2007). In another study with blood samples in the USA, mean Hg concentrations of pregnant women ( $0.69 \mu\text{g L}^{-1}$ ) were lower than those in non-pregnant ( $0.82 \mu\text{g L}^{-1}$ ) women (Jones *et al.* 2010). On the other hand, in a study with pregnant woman from Portugal, the average risk was below the unity (0.81); nevertheless, in 28% of the studied women, the exposure limit exceeded the unity (Nunes *et al.* 2014).

### **Contribution of the variables in MeHg ingestion**

The variables considered in the present risk characterization were  $IR_{food}$ ,  $BW$ ,  $D_{Days}$ , and  $C_{Food}$ . Based on rank correlations, in all cases,  $IR_{food}$  of fishery products was the main variable related to MeHg exposure (Figure 4); in contrast, body weight was negatively correlated

**Table 4.** Hazard quotient (HQ) values at different percentiles in surveyed population groups and percentage (%) of population in risk.

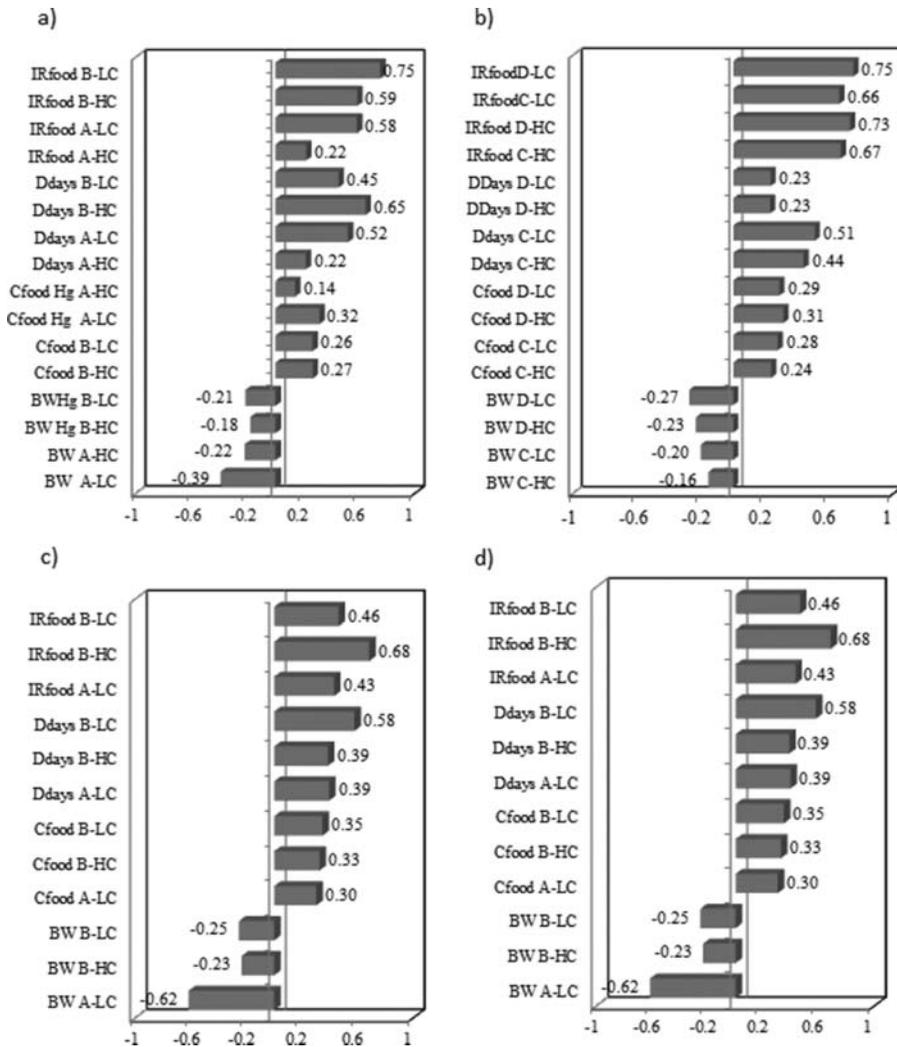
	N	Average	SD	P25	P50	P75	P95	P99	% in risk
General Population									
Low consumption									
A	8	0.59	1.87	0.18	0.33	0.65	1.77	3.89	13
B	15	0.52	1.23	0.14	0.25	0.49	1.66	3.76	10
C	38	0.33	0.65	0.14	0.22	0.38	0.89	1.66	4
D	47	0.18	0.19	0.09	0.14	0.22	0.46	0.83	No risk
High Consumption									
A	3	2.64	102.92	0.59	1.12	2.23	9.67	57.18	54
B	11	10.91	24.97	2.63	5.22	11.23	36.72	95.58	97
C	17	1.08	1.08	0.47	0.81	1.37	2.88	5.11	39
D	70	2.24	2.1	1.15	1.76	2.75	5.16	8.37	82
Fishing related population									
Low consumption									
A	9	1.55	17.49	0.70	1.06	1.70	4.06	9.33	51
B	5	0.35	0.27	0.22	0.29	0.40	0.72	1.20	1.5
C	18	0.33	0.54	0.11	0.19	0.35	0.99	2.29	5
D	17	0.20	0.19	0.12	0.16	0.23	0.48	0.93	No risk
High consumption									
B	8	1.88	1.68	0.98	1.41	2.23	4.87	8.51	71
C	27	1.89	2.19	0.84	1.27	2.07	5.22	11.30	65
D	77	2.48	3.23	1.07	1.75	2.94	6.69	13.10	78

A = Children A (3–10 years old); B = Children B (11–15 years old), C = Women C (women in childbearing age 16–40 years old), D = rest of population D (men > 16 years old and women > 41 years old).

with all the variables. After consumption, the body weight was an important variable related to Hg levels due to its capacity to distribute the contaminants in the body mass. In the case of children, who consume higher quantities of fishery products in relation to their body weight than adults, they can have a greater susceptibility to contaminants combined with less capability to detoxify them. In a similar study in French children, the authors found a negative correlation of body weight with other parameters like fish consumption rate (Morriset *et al.* 2013). In this context,  $IR_{food}$  and  $D_{Days}$  stand out as the most important factors in Hg exposure; therefore, even if the Hg content in fishery products could be relatively low (for instance, below Mexican regulation), the risk could be high if the frequency and quantity of consumption of fishery products are considerable.

### Perceptions of fish consumption

It is important to note that although FRP exhibited the highest  $IR_{food}$ , their HQs and percentage of population at risk were lower than those of GP. This is likely related to the differences in fishery products' preferences and affordability among the two populations, where the GP may have better economic means to acquire high priced fish, which in turn could have higher Hg content. In this context, the action level adopted by the Mexican government to prevent a potential public health issue (0.5 and 1 mg kg<sup>-1</sup> wet weight of MeHg depending on the fish species; NOM-242-SSA1-2009) is applicable for the general citizen, who has in many cases a lower fish consumption than people living in coastal communities characterized by high consumptions of fishery products. However, the importance of fish consumption for a healthy and nutritional status is undeniable; therefore, a communication effort in the form of fish advisories is desirable, considering local population characteristics, and its



**Figure 4.** Contribution of the variables in MeHg ingestion in the different groups of population. General population: (a) Children, (b) Adults. Fishing-related population: (c) Children, (d) Adults. A = Children A (3–10 years old); B = Children B (11–15 years old); C = Women C (women in childbearing age 16–40 years old); D = Rest of population D (men > 16 years old and women > 41 years old). LC = low consumption; HC = high consumption.

consumption habits and contaminant levels of fishery products. These advisories should prioritize the protection of sensitive and vulnerable population groups. In 2010, a joint group of experts (FAO/WHO 2010) consulted on the risk and benefits of fish consumption and emphasized that fish consumption lowers the risk of coronary heart disease mortality in adults and the risk of suboptimal neurodevelopment in offspring (due to maternal fish consumption). They associated the benefits to the occurrence of long-chain n-3 (or omega-3) polyunsaturated fatty acids (LCn3PUFAs), ultimately recommending the implementation of strategies to minimize risk and maximize benefits from consumption of fishery products. In this sense, WHO (2008) suggests communication strategies to send different messages to

different segments of a population according to their specific needs and risks. For example, while U.S. Food and Drug Administration (USFDA) sets a tolerance level of  $1 \text{ mg kg}^{-1}$ , the USEPA settles more conservative screening values (SV) for recreational ( $0.4 \text{ mg kg}^{-1}$ ) and subsistence fishers ( $0.049 \text{ mg kg}^{-1}$ ) due to differences in fish consumption (WHO 2008).

Considering that the probabilistic analysis was limited to only 18 food items with Hg data, a subestimation of Hg exposure is possible, especially in FRP. Levels of Hg in hair (GP 2.08 and FRP  $3.11 \mu\text{g g}^{-1}$ , unpublished data) also indicate that the actual exposure in the studied population could be of concern (according to World Health Organization (WHO) action level  $2 \mu\text{g g}^{-1}$ ; WHO 1990). Altogether, our results indicate the need of a monitoring program regarding MeHg in fishery products along with consumption patterns. Additionally, epidemiological studies suggested that fish-eating human populations may be exposed to sufficient Hg to cause significant developmental effects (Chan *et al.* 2003). Following the Minamata Convention (UNEP 2013), Mexico has been evaluating its capacity for Hg monitoring, legislation, and identification of contaminated sites. Therefore, new tools for safety assessments must be developed and further risk-benefit studies are desirable, as well as pollutants such as persistent organic contaminants should also be considered in an integral risk assessment.

## Conclusions

Food frequency surveys showed that fish consumption by Mazatlán people was well above than the Mexican national average. Although FRP presented a higher fish consumption rate than GP, MeHg exposure was higher in GP, likely due to differences in consumption patterns. In general, children showed the highest risk of MeHg exposure, being “children B” with high consumption of GP, the group exhibiting the maximum percentage of population at risk (97%).

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