

Total and Organic Mercury in Ten Fish Species for Human Consumption from the Mexican Pacific

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Abstract Total mercury and organic mercury were measured in ten fish species from the Mexican Pacific ocean to have a general view on the ratio of total mercury-organic mercury and potential implications on human health. Highest concentration of total mercury was recorded in muscle tissue of *Carcharhinus leucas* ($0.62 \mu\text{g g}^{-1}$ wet weight). Organic mercury was more concentrated in *Haemulon sexfasciatum* ($0.4 \mu\text{g g}^{-1}$ wet weight). Percentages of organic mercury ranged from 33 to 100%. Hazard indices associated to organic mercury and average fish consumption in Mexico ranged from 0.25 in *Lutjanus colorado* to 1.65 in *Haemulon sexfasciatum*.

Keywords Mercury (Hg) · Ichthyofauna · Risk assessment · Mexico

It has been widely recognized (Agah et al. 2007) that fish ingestion is the main pathway for total mercury (Hg) and organic Hg (mainly as methylmercury- MeHg) exposure in humans. A key feature related to variations in Hg content in fish is the feeding habit; in diverse studies (Pellegrini and Barghigiani 1989), it has been mentioned that individuals living in the proximity of surficial sediments and with a diet consisting mainly of fish have more elevated

levels of Hg and other metals than ichthyofauna from the pelagic environment that consume other type of food (such as crustaceans or molluscs). Mercury dynamics in fish from the marine environment is related to the ratio of MeHg and total Hg in muscle tissue; some authors have reported that from 85% to 97% of the total Hg is in the form of MeHg, but other researchers have found that MeHg may reach from 64% to 100% of the total amount of Hg (Agah et al. 2007). This variation in Hg accumulation is related to the processes that take part at the base of trophic chains. It was demonstrated that assimilation of inorganic and organic Hg by phytoplankton is not the same; while the organic Hg enters the cytoplasm, inorganic Hg is bound to the cell membrane; when plankton feeders capture their food, they digest the cytoplasmic content but eliminate the membrane of cytoplasmic cells where inorganic Hg is bound, this processes accounts for a differential accumulation of organic Hg with respect to the inorganic Hg (Mason et al. 1995).

Organic Hg accumulation is a key feature of the Hg cycle in the marine environment. It is well known that organic mercury, mainly as MeHg, is more toxic to humans than inorganic forms because of their capacity to cross biological membranes, high stability and potential of accumulation in animals and human food chain (Clarkson and Magos 2006). In this sense, it is necessary to know Hg concentrations in the edible portion of fish of human interest; moreover, the ratio of organic to inorganic Hg in fish of commercial value can help to advise population on the health risks associated to fish consumption. Health agencies as well as local and federal governments in some countries are concerned about Hg levels in fish for human consumption; in Mexico, the maximum permissible level for total Hg in fish and mollusks was set as $1.0 \mu\text{g g}^{-1}$ on a wet weight basis, whereas for MeHg this level is

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$0.5 \mu\text{g g}^{-1}$. Contrastingly, published studies related to the occurrence of total Hg in fish from Mexican waters in the Pacific Ocean are scarce; with respect to the organic form of Hg, no studies have been published in fish from the Mexican Pacific. The purpose of this study was to have a general view on the ratio of total Hg-organic Hg and the potential implications on the health of consumers.

Materials and Methods

In the present study, levels of total Hg and organic Hg were measured in muscle of nine fish species from the coastal waters of Mexico and one oceanic species from the eastern Pacific Ocean. Fish species considered in the present study are widely consumed by the Mexican population. Fish were collected by local fishermen from different sites in the eastern Pacific Ocean; a tuna specimen was collected and provided by a commercial vessel based in Mazatlán (SE Gulf of California; Fig. 1). Dates of collection, biometric (total weight and total length) data and other ecological information are provided in Table 1. Taxonomic identification of fish was made according to illustrated keys. The tissue of interest for the present study was muscle; dissection of the median dorsal portion was made by using a stainless steel scalpel. In order to avoid contamination of samples from manipulation and storage, glassware and other plastic utensils were previously washed according to

Moody and Lindstrom (1977). Muscle samples were placed in plastic bags for transportation at a low temperature (-5°C). Considering the size of the specimens and the amount of tissue required for Hg determinations, a total of 18 individual samples were analyzed. Muscle samples were lyophilized for 72 h (133×10^{-3} mBar and -49°C) then manually ground with an agate mortar and pestle. Portions (0.25 g) of powdered and homogenized muscle samples were used for total Hg and organic Hg analysis.

For total Hg analysis, samples were pre-digested with concentrated nitric acid (5 mL; trace metal grade) overnight using Teflon vessels (Savillex). Digestion was made by using capped Teflon vessels and a hot plate at 120°C for 3 h. Digested samples were stored in polyethylene containers for further analysis. Analyses were carried out by reducing mercury compounds in solution samples using SnCl_2 (Loring and Rantala 1995); measurements were made by cold vapour atomic absorption spectrophotometry (CV-AAS) with a Varian SpectraAA220 equipment. For organic mercury determinations, four steps were followed according to (Barregard et al. 1993): (a) acid extraction with HCl 6 M; (b) separation of inorganic Hg by ionic exchange in glass columns filled with a Dowex resin (Cl-form; 100–200 mesh size); (c) conversion of organic Hg to Hg^{+2} by ultraviolet radiation (100 w lamp) during 48 h; and (d) detection by CV-AAS. In order to minimize matrix interferences with organic Hg (Sanz-Landaluze et al. 2004), the standard addition method was used. Additionally, antifoaming silicon (Merck) was used to avoid the saponification of fatty acids in fish tissues (Gómez-Ariza et al. 2004).

Total Hg and organic Hg concentrations are expressed as $\mu\text{g g}^{-1}$ on a wet weight basis; precision and accuracy of the analytical method were assessed by using certified reference material (NIST-2977); calculated total and organic Hg fell (recoveries 95% and 99%, respectively) within certified values. The variation coefficients were below 10%. Limit of detection and of quantification for total Hg and organic Hg were 0.025 and $0.125 \mu\text{g g}^{-1}$ (wet weight) respectively. To check for contamination, blanks were run with every batch of samples. Hazard index (HI) for organic Hg was estimated according to (Newman and Unger 2002): $\text{HI} = \text{E}/\text{RfD}$, where E is the exposure level or intake of organic mercury, and RfD is the reference dose ($0.1 \mu\text{g kg}^{-1} \text{day}^{-1}$). The exposure level (E) is calculated as $\text{E} = \text{C} \times \text{I}/\text{W}$; where C is the concentration of organic Hg in fish, I is the ingestion rate of fish (25 g day^{-1}) and W is the weight of an average adult (70 kg).

Results and Discussion

From Table 1, it can be seen that fish from the different sites belonged to different species, such situation is

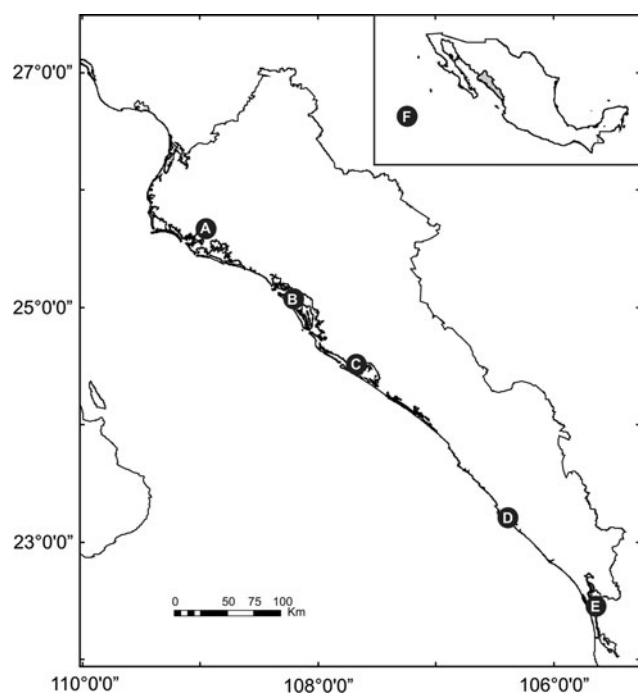


Fig. 1 Collection sites of fish in the Mexican Pacific: *a* Topolobampo, *b* Santa María, *c* Altata-Ensenada del Pabellón, *d* Urías; *e* Teacapán; *f* Open ocean

Table 1 Dates and sites of collection, biological information and levels of Hg (in $\mu\text{g g}^{-1}$ wet weight) in muscle tissue of studied fish

Scientific name (common name)	N	Site code ^a	Weight (g)	Length (cm)	Date of collection	Habitat	Feeding mode	Total Hg	Organic Hg	%Organic Hg
<i>Caranx caninus</i> (Pacific crevalle jack)	3	A	45–72	14–18	March/2004	BP	Ca	0.20 ± 0.04	0.20 ± 0.07	100
<i>Nematistius pectoralis</i> (Roosterfish)	4	A	139–173	23–25	March/2004	P	Ca	0.46 ± 0.02	0.32 ± 0.05	69
<i>Roncador steurnsii</i> (Spotfin croaker)	1	A	88	19	March/2004	De	Ca	0.27 ± 0.04	0.26 ± 0.09	96
<i>Haemulon sexfasciatum</i> (Greybar grunt)	3	B	77–134	18–22	November/2003	De	Ca	0.60 ± 0.02	0.40 ± 0.05	66
<i>Trachinotus patiensis</i> (Pompano)	1	B	285	30	November/2003	BP	Ca	0.27 ± 0.03	0.24 ± 0.03	89
<i>Elops affinis</i> (Pacific ladyfish)	1	B	251	38	November/2003	P	Ca	0.34 ± 0.03	0.34 ± 0.09	100
<i>Carcharhinus leucas</i> (Bull shark)	1	C	11,000	106	December/2003	P	Ca	0.62 ± 0.20	0.27 ± 0.05	44
<i>Lutjanus colorado</i> (Colorado snapper)	1	D	372	29	June/2003	De	Ca	0.18 ± 0.17	0.06 ± 0.02	33
<i>Gerres cinereus</i> (Yellowfin mojarra)	2	E	245–262	22–23	January/2004	De	O	0.29 ± 0.08	0.23 ± 0.04	79
<i>Thunnus albacares</i> (Yellowfin tuna)	1	F	10 000	87	July/2005	P	Ca	0.20 ± 0.17	0.20 ± 0.01	100

^a See Fig. 1 for location of collection sites; A, Topolobampo; B, Santa María; C, Altata-Ensenada del Pabellón; D, Uriás; E, Teacapán; F, open ocean
 Ca carnivorous, O omnivorous, De demersal, P pelagic, BP benthopelagic, N number of individuals

common in tropical and subtropical latitudes where biodiversity is elevated. Fish habitat is important in terms of the prevailing characteristics of the surrounding environment; fish collected here corresponded to pelagic (40%), demersal (40%) and benthopelagic (20%) habitats. In relation to the feeding behavior, with the exception of the yellowfin mojarra *Gerres cinereus*, studied species corresponded to carnivores; since diet is considered as a key factor in relation to Hg uptake in fish, it is important to consider the feeding mode in order to make a more complete interpretation of results.

Highest concentration of total Hg was recorded in muscle tissue of the bull shark *Carcharhinus leucas* ($0.62 \mu\text{g g}^{-1}$) from site C (Table 1); it is probable that the feeding mode (carnivorous), combined with the degree of anthropogenic development in that lagoonal system (intensive agriculture, urban developments, aquacultural ponds and sugar cane facilities) may account for such pattern. Organic Hg was more concentrated in muscle tissue of the carnivorous fish *Haemulon sexfasciatum* ($0.4 \mu\text{g g}^{-1}$) from site B, this area is also impacted (shrimp aquaculture, urban discharges) but in a lesser extent than site C. The percentage of organic Hg with respect to total Hg was highly variable (from 33% to 100%) in our study; in other regions where fish have been analyzed (Magalhães et al. 2007; Carbonell et al. 2009) it has been found that the percentage of organic Hg with respect to total Hg is over 80%. Other researchers (Holsbeek et al. 1997; Agah et al. 2007) have reported lower ratios (from 19% to 64%) of total to organic Hg. In this context, it has been proposed that fish can be grouped into three categories, depending on the patterns of Hg accumulation (Holsbeek et al. 1997): type I, when the concentration of organic Hg increases with the length of fish and inorganic fraction stays at a constant low level; type II with a low and constant organic Hg fraction but the inorganic fraction increases with length of fish; and type III, when both Hg fractions increase with fish length. In the present study, it was not possible to define a pattern of Hg variation with length of specimens due to the small number of samples, but 60% of the ichthyofauna had an elevated percentage of the organic Hg fraction and a low concentration of inorganic Hg as indicative of a type I behavior. In this sense, it has been concluded that this pattern is generally considered as the normal situation.

For comparative purposes, levels of total Hg and organic Hg in muscle tissue of fish from elsewhere are presented in Table 2. Highest levels of total Hg and organic Hg corresponded to the common mora *Mora moro* (3.70 and $2.96 \mu\text{g g}^{-1}$) and the ghostshark *Chimaera monstrosa* (3.14 and $2.67 \mu\text{g g}^{-1}$) from the Mediterranean; the reasons for such elevated Hg concentration are related to the characteristics of the deep environments where these species live (Johnson and Stevens 2000), combined with an

Table 2 Levels of total Hg and organic Hg (in $\mu\text{g g}^{-1}$ wet weight) in muscle tissue of marine fish from diverse sites

Species	Common name	Total Hg	Organic Hg	Organic Hg (%)	Site	References
<i>Chimaera monstrosa</i>	Ghostshark	3.14	2.67	83	Mediterranean Sea	Storelli et al. (2002)
<i>Saurida tumbil</i>	Greater lizard fish	0.04	0.04	100	Persian Gulf	Agah et al. (2007)
<i>Hilsa kelee</i>	Shad	0.021	0.004	20	Bangladesh	Holsbeek et al. (1997)
<i>Ilisha indica</i>	Jewelled shad	0.015	0.004	19	Bangladesh	Holsbeek et al. (1997)
<i>Ilisha filigera</i>	Jewelled shad	0.016	0.004	27	Bangladesh	Holsbeek et al. (1997)
<i>Ageneiosus caucanus</i>	Flatnose catfish	0.51	0.49	95	Mojana region, Colombia	Marrugo-Negrete et al. (2008)
<i>Tripotheus magdalenae</i>	Arenca	0.34	0.31	93	Mojana region, Colombia	Marrugo-Negrete et al. (2008)
<i>Lepidopus caudatus</i>	Silver scabbard fish	1.44	1.33	89	The Azores, Portugal	Magalhães et al. (2007)
<i>Conger conger</i>	European conger	1.86	1.69	91	The Azores, Portugal	Magalhães et al. (2007)
<i>Mora moro</i>	Common mora	3.70	2.96	84	The Azores, Portugal	Magalhães et al. (2007)
<i>Haemulon sexfasciatum</i>	Greybar grunt	0.60	0.40	66	Santa María, Mexico	This study
<i>Carcharhinus leucas</i>	Bull shark	0.62	0.27	44	AEP, Mexico	This study

elevated trophic position of these predators (Kress et al. 1998). The species from the Mexican Pacific showed low total Hg (around $0.6 \mu\text{g g}^{-1}$) and organic Hg (from 0.27 to $0.4 \mu\text{g g}^{-1}$) levels, such concentrations are in the same order of magnitude to those reported in fish from the Mojana region (Colombia), an area impacted by gold mining activities. In relation to the percentages of organic Hg, compared data can be grouped into two groups, the ones with levels above 80%, which are considered as normal (Bloom 1992), and a smaller group comprising the cases where organic Hg levels were below 70%. In muscle tissue of fish from the present study, organic Hg values ranged from 100% to 33%, six fish species had organic Hg percentages from 79 to 100, and four fish species were below 70 (Table 1).

Hazard indices (HI) associated to levels of organic Hg and average fish consumption by the Mexican population are presented in Table 3. This index is useful for estimating the level of exposure to a single substance (E); if HI is above unity, there may be potential health effects (i.e. if the

reference dose (RfD) is lower than calculated E, the values are above unity). Analyzed ichthyofauna presented HI that ranged from 0.25 in *L. colorado* to 1.65 in *H. sexfasciatum*; in 50% of the studied fish, values were higher than unity, it implies that there is a potential health risk from the consumption of these fish species according to the rate of fish consumption in Mexico. It is worth mentioning that RfD ($0.1 \mu\text{g kg}^{-1} \text{day}^{-1}$) developed by the US Environmental Protection Agency (EPA 1989) corresponds to methylmercury (MeHg), but considering that most organic Hg is in the form of MeHg such value was used here for HI calculations. In comparison with other studies, HI values obtained here were below results presented in the Mojana region, Colombia (HI from 1.5 to 5.9) where daily fish consumption is elevated (120 g per capita) and gold-mining activities exist (Marrugo-Negrete et al. 2008); but in comparison to a study in Tianjin (China) (daily fish consumption ranges from 38 to $57.5 \text{ g per capita}$) where HI were from 0.03 to 0.40 (Wang et al. 2005), values estimated here were more elevated.

Though Se was not included in this study, Hg toxicity should be considered as a multifactor issue; in this context, Se in marine fish has been shown to confer protective effects when elevated levels of MeHg are added to diets to induce toxicity in vertebrates (Ganter and Sunde 2007). From this statement, health risk posed by Hg exposure through fish consumption should be made considering the concurrent levels of Se since it could influence the Hg bioaccessible fraction (Kaneko and Ralston 2007), i.e., the sole presence of mercury does not imply necessarily a direct toxic effect on humans. More work is necessary on the consumption rates in different sectors of the Mexican population in order to estimate HI for single and several metals as well as more precise information related to consumption patterns since fish consumption is highly variable. On the other hand, in this study the number of

Table 3 Hazard indices (HI) for organic Hg of analyzed ichthyofauna from the Mexican Pacific

Species	HI
<i>Caranx caninus</i>	0.83
<i>Nematistius pectoralis</i>	1.30
<i>Roncador stearnsii</i>	1.13
<i>Haemulon sexfasciatum</i>	1.65
<i>Trachinotus paitensis</i>	0.99
<i>Elops affinis</i>	1.38
<i>Carcharhinus leucas</i>	1.15
<i>Lutjanus Colorado</i>	0.25
<i>Gerres cinereus</i>	0.94
<i>Thunnus albacores</i>	0.84

individuals was relatively low (1–4) that a sound conclusion is difficult to establish, therefore more research is needed.

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