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Health Risk Associated to Dietary Intake of Mercury in Selected Coastal Areas of Mexico

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Abstract A hazard quotient (HQ) was evaluated for Mexican population considering Hg levels and consumption rates. Fish and shrimp were caught in selected coastal areas. HQ's ranged from 0.004 to 0.01 in shrimp; from 0.004 to 1.04 in fish from NW Mexico; and from 0.02 to 0.19 in fish from the Gulf of Mexico. Highest HQ in fish were found in carnivorous fish *Caranx caninus* (HQ = 0.71) and *Sphyrna lewini* (HQ = 1.04) from NW Mexico. A more accurate study should be made considering organic Hg, contribution of Hg from other sources, and consumption rates according to age, sex and economic status.

Keywords Risk assessment · Fish · Shrimp · Hazard quotient · Mercury

Mercury is supplied to the environment through natural processes (mineral deposits, volcanoes, forest fires, oceanic emissions and crust degassing) and by anthropogenic activities as cement kilns, roasting of sulfide ores for production of sulfuric acid and smelters where sulfide ores are processed (i.e., in the production of gold, copper, iron, lead and zinc) (Hylander and Meili 2003). An average global estimation of 4,500 metric tons per year of anthropogenic emissions was made by Lindqvist et al. (1991); these figures indicate that current mercury deposition is a factor of 2–5 times greater than preindustrial deposition measurements. From the toxicological point of view, accumulation of Hg in biota is the most important feature of the marine Hg cycle (Fitzgerald et al. 2007) perhaps because it causes irreversible damage to aquatic and terrestrial biota. Health effects from Hg exposure consists mainly on the inhibition of neurological and cardiovascular development in children (Murata et al. 2007), and alteration of the cardiovascular health of adults.

Potential health risks due to Hg levels, as well as a combination of trace metals, can be estimated by non-cancer risk assessment methods by using the hazard quotient (HQ) and the hazard index (HI) set by Environmental Protection Agency of the United States (US EPA 1989); when using these indices it is assumed that there is a level of metal exposure (the reference dose, RfD) below which it is unlikely, even for sensitive populations, to experience adverse health effects (Marrugo-Negrete et al. 2008). The term RfD is the concentration of a given pollutant that can be ingested over a lifetime without health risk; for Hg, the value is 0.5 μg kg^{-1} day^{-1} (US EPA 2000). If HI and HQ are above unity, there is concern for potential health effects. Obviously, if the RfD for a given element is very low but the general population has a low consumption of fish and seafood, the health risk is minimum; in this context, it is necessary to have information related to the consumption of fishery products by different strata of the population. In Mexico, fishery products are not consumed very abundantly; on the average, fish is consumed at a rate of 9.01 kg $person^{-1}$ $year^{-1}$ (equivalent to 25 g $person^{-1}$ day^{-1}) and shrimp are consumed even less, around 1.49 kg $person^{-1}$ $year^{-1}$ (equivalent to 4.1 g $person^{-1}$ day^{-1}) (Ruelas-Inzunza et al. 2010). From the above picture, the

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potential health risk to Mexican population was assessed in selected coastal areas of the country, considering the consumption patterns of shrimp and fish as well as the concentrations of Hg in comparison to reference doses established by the US EPA.

Materials and Methods

Collection of specimens was made in Guaymas (GH) harbor (fish) and Sinaloa (SIN) coastal zone (fish and shrimp) in the NW coast of Mexico; in the Gulf of Mexico (SE coast of Mexico), fish were caught in Coatzacoalcos Estuary (CE) (Fig. 1). Potential sources of metal pollution in GH comprise oil residues from transport operations, domestic effluents from the city of Guaymas (population > 150,000), effluents from a cement industry and cooling waters from an electrical plant (Green-Ruiz 2000). In SIN state, approximately 2.5 million people live, the littoral zone is 640 km long; estuaries and coastal lagoons comprise 221,600 ha; potential sources of pollution in the zone are fisheries, agriculture, shrimp farming, urban sewage and shipyards. Coatzacoalcos estuary is close to the main oil resources in the Gulf of Mexico where industrial development has occurred in the past 40 years.

Shrimp and fish were collected by local fishermen. Data of species as well as date of sampling are provided in Table 1. Shrimp specimens were identified according to Hendrickx (1995), measured and weighed, more details on the sampling procedures can be found in Ruelas-Inzunza et al. (2004). Fish were collected by local fishermen; taxonomic identification was made by using illustrated keys (Allen et al. 1995). Glassware and plastic utensils used for sample preparation were previously washed (3 days immersed in HCl 2 M; rinsing with milliQ water; 3 days immersed in HNO3 2 M; final rinsing with milliQ water) according to Moody and Lindstrom (1977). Samples were freeze-dried for 72 h (−49°C; 133×10^{-3} mbar) then ground in an agate mortar with pestle. Powdered samples were acid digested (5 mL of quartz distilled concentrated nitric acid) using a microwave digestion system (CEM-MDS-2000). Digested samples were stored in polyethylene containers for further analysis. Analyses of Hg were made by cold vapor atomic absorption spectrophotometry (CVAAS). Precision and accuracy of the method were assessed by using reference material MA-B-3/TM (IAEA 1987); a satisfactory agreement between the analytical results (0.60 ± 0.05 μg g^{-1} dry weight) and the certified values (0.54 ± 0.07 μg g^{-1} dry weight) was obtained. More details on this matter may be found in Ruelas-Inzunza et al. (2009) and Ruelas-Inzunza and Páez-Osuna (2005). Concentrations of Hg are reported in μg g^{-1} on a wet weight basis.

Hazard quotient (HQ) was estimated according to Newman and Unger (2002): HQ = E/RfD, where E is the

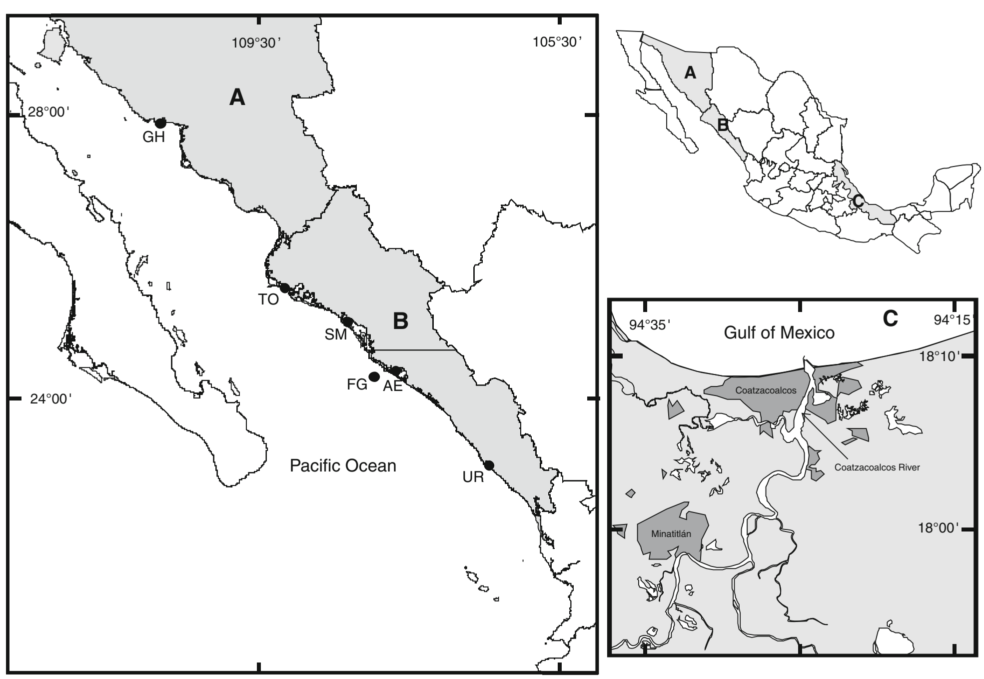


Fig. 1 Location of coastal areas where fish and shrimp were collected (*A* Sonora state, *B* Sinaloa state, *C* Veracruz state, *GH* Guaymas harbor, *TO* Topolobampo, *SM* Santa María, *AE* Altata-Ensenada del Pabellón, *FG* fishing ground, *UR* Urías)

Table 1 Scientific names, common names, feeding behavior and dates of collection of fish and shrimp from selected coastal areas in Mexico

Species	Common name	Feeding habit	Date of collection	Site
Fish				
Opisthonema libertate	Pacific thread herring	H	Apr 1998	GH
Cathorops fuerthii	Catfish	C	Apr 1998	GH
Mugil cephalus	Striped mullet	O	Apr 1998	GH
Seriola lalandi	Yellowtail	C	Apr 1998	GH
Galeichthys peruvianus	Catfish	C	Dec 1998–Jan 1999	AP
Lutjanus colorado	Colorado snapper	C	Dec 1998–Jan 1999	AP
Cynoscion xanthulus	Orangemouth corvine	C	Dec 1998–Jan 1999	AP
Mugil cephalus	Striped mullet	O	Dec 1998–Jan 1999	AP
Carcharhinus leucas	Bull shark	C	Dec 1998–Jan 1999	AP
Sphyrna lewini	Scalloped hammerhead	C	Dec 1998–Jan 1999	AP
Pomadasys leuciscus	Gray grunt	C	Mar 2004	TO
Scomberomorus sierra	Spanish mackerel	C	Mar 2004	TO
Diapterus peruvianus	Peruvian mojarra	C	Mar 2004	TO
Roncador stearnsii	Spotfin croaker	C	Mar 2004	TO
Oligoplites saurus	Leatherjack	C	Mar 2004	TO
Elops affinis	Pacific ladyfish	C	Mar 2004	TO
Mugil cephalus	Striped mullet	O	Mar 2004	TO
Centropomus armatus	Longspine snook	C	Mar 2004	TO
Haemulopsis axillaris	Yellowstripe grunt	C	Mar 2004	TO
Nematistius pectoralis	Rooster fish	C	Mar 2004	TO
Paralichthys woolmani	Speckled flounder	C	Mar 2004	TO
Caranx caninus	Pacific crevalle jack	C	Mar 2004	TO
Selar crumenophthalmus	Bigeye scad	O	Mar 2004	TO
Arius platypogon	Slender-spined catfish	O	Mar 2004	TO
Centropomus nigrescens	Black snook	C	Mar 2004	TO
Haemulon sexfasciatum	Graybar grunt	C	Dec 2003	SM
Mugil curema	White mullet	O	Dec 2003	SM
Trachionotus paitensis	Paloma pompano	C	Dec 2003	SM
Elops affinis	Pacific ladyfish	C	Dec 2003	SM
Gerres cinereus	Yellowfin mojarra	O	Dec 2003	SM
Centropomus medius	Blackfin snook	C	Dec 2003	SM
Haemulopsis axillaris	Yellowstripe grunt	C	Dec 2003	SM
Sphoeroides annulatus	Bullseye puffer	C	Dec 2003	SM
Mugil cephalus	Striped mullet	O	Feb 2004	AP
Caulolatilus princeps	Ocean whitefish	C	Feb 2004	AP
Mugil curema	White mullet	O	Jun 2003	UR
Lutjanus colorado	Colorado snapper	C	Jun 2003	UR
Mugil curema	White mullet	O	Jul 2003	FG
Diapterus peruvianus	Peruvian mojarra	C	Jul 2003	FG
Sphoeroides annulatus	Bullseye puffer	C	Jul 2003	FG
Pomadasys branickii	Sand grunt	C	Jul 2003	FG
Eucinostomus gracilis	Slender mojarra	C	Jul 2003	FG
Oreochromis sp.	Tilapia	O	May 2005	CE
Gobiomorus polylepis	Finescale sleeper	C	Jan 2006	CE
Gerres cinereus	Yellowfin mojarra	O	Sep 2005	CE
Centropomus viridis	White snook	C	Sep 2005	CE
Lepisosteus osseus	Longnose gar	C	Jan 2006	CE

Table 1 continued

Species	Common name	Feeding habit	Date of collection	Site
Shrimp				
Litopenaeus stylirostris	Blue shrimp	O	Dec 1998–Jan 1999	AP
Litopenaeus vannamei	White shrimp	O	Dec 1998–Jan 1999	AP
Farfantepenaeus brevirostris	Crystal shrimp	O	Dec 1998–Jan 1999	AP
F. californiensis	Brown shrimp	O	Dec 1998–Jan 1999	AP
Xiphopenaeus kroyeri	Pacific seabob	O	Dec 1998–Jan 1999	AP

GH Guaymas harbor, *AP* Altata-Ensenada del Pabellón, *TO* Topolobampo, *SM* Santa María, *UR* Urías, *FG* fishing ground, *CE* Coatzacoalcos Estuary

exposure level or intake of total Hg, and RfD is the reference dose for total Hg (0.5 μg kg^{-1} day^{-1}). The exposure level (E) is calculated as E = C × I/W; where C is the concentration of total Hg in fish or shrimp, I is the ingestion rate of fish (25 g day^{-1}) and shrimp (4.1 g day^{-1}) in Mexico and W is the weight of an average adult (70 kg). Conversion of Hg concentrations from dry weight (Hg_{dw}) to fresh weight (Hg_{fw}) were made according to the equation: $Hg_{fw} = Hg_{dw}$ * (100–% humidity)/100 (Magalhães et al. 2007), using an average of 70% of humidity. Comparisons of average Hg concentrations and HQ among fish from the different sites were made by a one-way ANOVA; statistical differences between HQ's of fish and shrimp were defined by a t-Student test using GraphPad Prism 4.0 (Graph Pad Software, San Diego, CA).

Results and Discussion

Collected fish from the studied areas have different feeding behaviours, carnivore was the most frequent mode (74%) followed by omnivores (24%) and herbivores (2%) (Table 1). It is known that Hg is susceptible to biomagnification (Dietz et al. 2000) so the feeding habit is an important factor related to expected Hg concentrations in fish, i.e., fish of a higher trophic position are more likely to have increased Hg levels in comparison to fish of lower trophic levels; in the present study, fish with the most elevated Hg concentrations were carnivores. In relation to Hg levels in muscle tissue of analyzed biota from the different sites and comparison to legal limits in fishery products for human consumption, average concentrations of Hg comprising biota for every site are presented in Table 2. Average Hg concentrations were converted to fresh weight basis since this is the most common way to present information in the international legislation. Comparison of averaged Hg concentration among fish from the studied sites were not significantly ($p > 0.05$) different. Averaged Hg concentrations in muscle tissue of fish above the Mexican legislation (1.0 μg g^{-1} wet weight) were found only in fish from Sinaloa state (SIN). Concentration of Hg in shrimp species were far below national and international legislations. As mentioned before, legal limits of trace metals for diverse fishery products are variable depending on the country (Nauen 1983); for Hg, the most common legal limit is 1 μg g^{-1} wet weight but some countries (Canada and Brazil) have lower thresholds (0.5 μg g^{-1} wet weight). Given the heterogeneity of criteria for setting the legal limits in the different countries, special care should be taken with species whose Hg levels were below the Mexican legislation but above the Belgian regulation; such is the case of diverse fish species from Sinaloa state.

Table 2 Average concentrations (μg g^{-1}, wet weight) of Hg in fish and shrimp and comparison to maximum permissible levels (in μg g^{-1} wet weight) in fishery products according to national and international legislations

State	Site	Hg (μg g^{-1})	Samples above MPL[a] (%)	Samples above MPL[b] (%)
Fish				
Sonora	GH	0.11 ± 0.09	0	0
Sinaloa	AP	0.31 ± 0.48	12.5	37.5
	TO	0.31 ± 0.23	6.7	40
	SM	0.29 ± 0.14	0	50
	UR	0.06 ± 0.05	0	0
	FG	0.25 ± 0.1	0	40
Veracruz	CE	0.14 ± 0.11	0	0
Shrimp				
Sinaloa	AEP	0.06 ± 0.02	0	0

MPL maximum permissible levels, [a] Mexican legislation (0.1 μg g^{-1}), [b] Belgian legislation (0.3 μg g^{-1}), *GH* Guaymas harbor, *AP* Altata-Ensenada del Pabellón, *TO* Topolobampo, *SM* Santa María, *UR* Urías, *FG* fishing ground, *CE* Coatzacoalcos estuary

The detailed information on Hg levels in analyzed biota is depicted in Fig. 2. Considering all the analyzed fish species (47), only two species (4.2%) showed Hg levels that were equal or above the Mexican legislation but fourteen species (30%) had levels equal or above legal

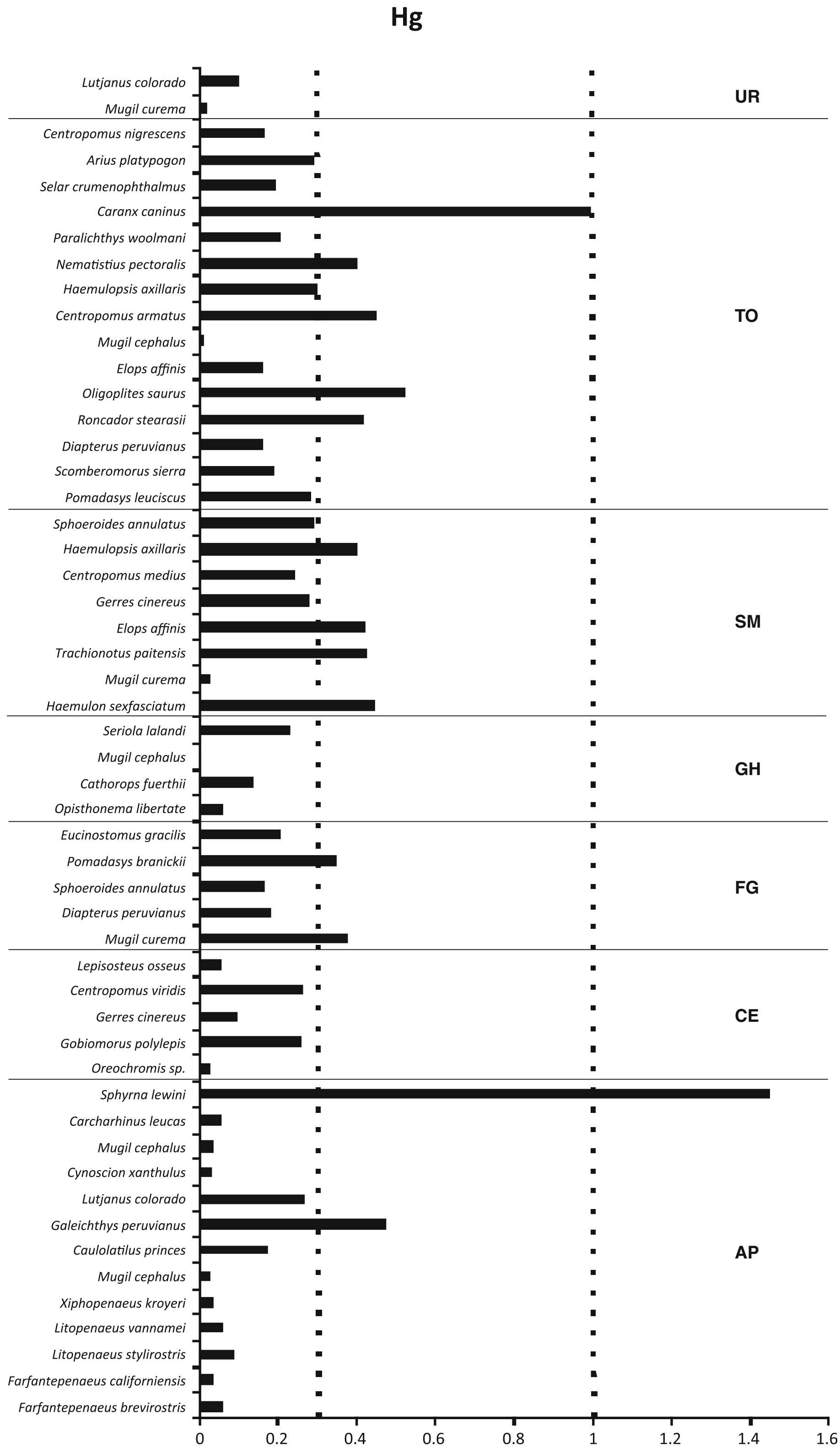


Fig. 2 Concentrations of Hg (in μg g^{-1}, wet weight) in muscle of fish and shrimp from selected coastal areas in Mexico (*UR* Urías, *TO* Topolobampo, *SM* Santa María, *GH* Guaymas harbor, *FG* fishing ground, *CE* Coatzacoalcos estuary, *AP* Altata-Ensenada del Pabellón)

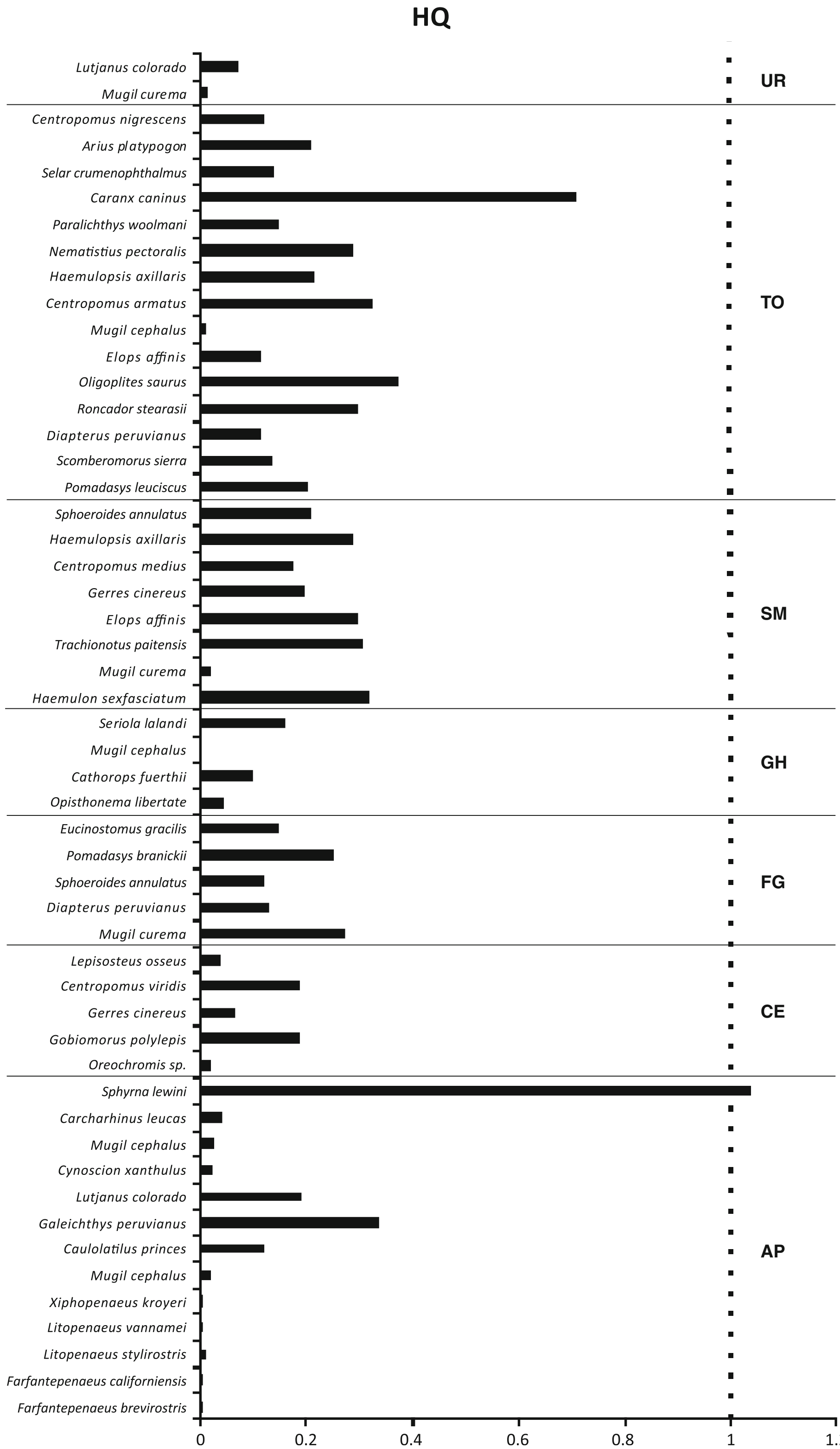
limits considered in the Belgian legislation. The scalloped hammerhead *Sphyrna lewini* from AP had 1.452 μg g^{-1} wet weight and the Pacific crevalle jack *Caranx caninus* from TO had 1.0 μg g^{-1} wet weight, both species had Hg levels above the national legislation. Considering the Belgian legislation, only fish from SIN had Hg levels (in μg g^{-1} wet weight) above or equal to the established limit: *S. lewini* and *Galeichthys peruvianus* (0.474) from AP; *C. caninus*, *Roncador stearasii* (0.407), *Oligoplites saurus* (0.522), *Centropomus armatus* (0.453), *Haemulopsis axillaris* (0.303) and *Nematistius pectoralis* (0.402) from TO; *Trachionotus paitensis* (0.426), *Elops affinis* (0.42), *H. axillaris* (0.402) and *Haemulon sexfasciatum* (0.447) from SM; *Mugil curema* (0.381) and *Pomadasys branickii* (0.351) from FG. None of the shrimp species had Hg levels above the maximum permissible limits in fishery products for human consumption.

Considering the sites where fish had the most elevated concentrations (i.e. TO and AP in the State of Sinaloa), main anthropogenic activities might be related to the occurrence of such levels of Hg. In AP the main activities are related to agriculture, shrimp farming, urban sewage and fisheries (Páez-Osuna et al. 2002) whereas in TO potential sources of metals are related to industrial activity as well as agriculture and shrimp farming. In addition, the feeding habit of the fish species might also account for such levels, i.e. carnivore fish showed the highest Hg levels. It has been established that comparisons of metal concentrations should be made between organisms of the same species (Rainbow and Phillips 1991); from published studies, it was found that in *Sphyrna lewini* from AP (present study) Hg levels (1.452 μg g^{-1} wet weight) were comparable to levels (1.08) reported in organisms of the same species from other sites in the Gulf of California (García-Hernández et al. 2007) but an order of magnitude higher than in specimens (0.76 μg g^{-1}) from Papua, New Guinea (Powell and Powell 2001). In the case of *Mugil curema*, organisms from FG had levels (0.381 μg g^{-1} wet weight) above specimens from SM and UR (0.027 and 0.018 respectively) and *M. curema* (0.048) from Cumaná, Venezuela (Shrestha et al. 1988).

Hazard quotients (HQ) for every fish and shrimp species are shown in Fig. 3. Considering all the collected biota, values of HQ in fish were significantly higher ($p < 0.05$) than in shrimp. Comparisons of HQ among fish from the different sites were not significantly ($p > 0.05$) different. Since HQ are estimated on the basis of the consumption rates of fishery products in a certain population, it is important to highlight such figures; from an average consumption of fish and shrimp (9.01 and 1.49 kg *per capita* per year) in Mexico, it turns out that the daily consumption *per capita* is 25 g for fish and 4.1 g for shrimp; such figures are lower than the world average fish consumption of 63 g per day (WWF 2000) and a daily fish consumption of 120 g in the Mojana region, Colombia (Marrugo-Negrete et al. 2008). Unfortunately, information concerning detailed patterns of consumption of fish and shrimp species by the Mexican population is non-existing. The other factor directly involved in HQ fluctuation is the Hg concentration in the edible portion of fishery products; in this respect it is necessary to generate information regarding patterns of consumption, especially in fishermen, anglers and their families, since their fish intake is above the average in Mexico. We estimated that fishermen and their families consume two or three times more fishery products in comparison to the average; consequently, this sector of the Mexican population is of concern. With the exception of *Sphyrna lewini* (HQ = 1.04), the rest of the analyzed biota showed HQ below unity; therefore, it can be mentioned that the daily intake of Hg through the ingestion of the collected fish and shrimp are not likely to affect health of Mexican consumers; moreover, addition of individual HQ's (those of fish and shrimp) were also below one, it means that the combined effect of Hg ingestion from fish and shrimp are not likely to pose adverse effects on consumers. This is particularly important for pregnant women because it has been documented that more than 90% of mercury in fish is present as methylmercury (Morel et al. 1998) and this Hg species accumulates in the brain, especially in developing fetuses. However, another factor that is worth mentioning is that even sea food products are the main sources of Hg, other foodstuffs like cereals, beans, eggs and vegetables also contribute to the total load of Hg (Zheng et al. 2007); from this assumption, it is necessary to keep in mind that even if HQ's are lower than unity in most of the analyzed fishery products, contribution from other dietary items may be important in terms of the potential health risk of consumers. The sequence of averaged HQ in the analyzed biota followed the order carnivorous fish > omnivorous fish > herbivorous fish > shrimp, from this trend, it can be said that frequent fish consumers should be cautious when consuming elevated amounts of carnivorous fish, especially top predators like sharks.

Comparison of hazard quotients (HQ) associated to dietary intake of Hg through consumption of fish and other food items is presented in Table 3. In general, HQ's associated to fish were more elevated than values associated to wheat and shrimp consumption. HQ's varied by three orders of magnitude; highest values were estimated for adults with an elevated fish consumption (120 g) in the Mojana region (Colombia), an area where gold-mining is a common activity. On the other hand, lowest HQ's were calculated for shrimp from Sinaloa, Mexico (this study); such low values were derived from a low consumption of shrimp (4.1 g) in Mexico and low Hg concentrations in the edible portion. The other study with a low HQ value was found for a low consumption (17.4 g) of sea products in children from Huludao city (China).

Fig. 3 Hazard quotients (HQ) in fish and shrimp species from selected coastal areas in Mexico (*UR* Urías, *TO* Topolobampo, *SM* Santa María, *GH* Guaymas harbor, *FG* fishing ground, *CE* Coatzacoalcos estuary, *AP* Altata-Ensenada del Pabellón)



Considering Hg concentrations and average per capita consumption of fish and shrimp in the Mexican population, it can be concluded that only the carnivorous fish *Caranx caninus* and *Sphyrna lewini* represent a potential health risk for consumers in the NW coast of Mexico where these fish were collected.

Table 3 Comparison of hazard quotients (HQ) associated to dietary intake of Hg in different regions of the world

Dietary item	Site	HQ	Daily consumption and population considered
Sea products	Huludao city, China[a]	0.022	55 g, adults
Sea products	Huludao city, China[a]	0.012	17.4 g, children
Fish	Bei Chen, China[b]	0.10	57.5 g, adults
Fish	Bei Chen, China[b]	0.40	92.6 g, children
Fish	Mojana region, Colombia[c]	4.0	120 g, adults
Wheat grain	Kunshan, China[d]	0.026	144 g, rural adults
Wheat grain	Kunshan, China[d]	0.026	47.8 g, rural children
Fish	Sinaloa, México[e]	0.2062	25 g, adults
Fish	Sonora, México[e]	0.078	25 g, adults
Fish	Veracruz, México[e]	0.1020	25 g, adults
Shrimp	Sinaloa, México[e]	0.0068	4.1 g, adults

[a] Zheng et al. (2007)

[b] Wang et al. (2005)

[c] Marrugo-Negrete et al. (2008)

[d] Huang et al. (2008)

[e] This study

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