Bull Environ Contam Toxicol
DOI 10.1007/s00128-016-1811-x

Total Mercury in *Mugil* spp and *Eugerres axillaris* of a Subtropical Lagoon of NW Mexico

M. G. Frías-Espericueta[1] · A. Vargas-Jiménez[2] · J. Ruelas-Inzunza[3] · J. I. Osuna-López[1] · M. Aguilar-Juárez[1] · J. C. Bautista-Covarrubias[4] · D. Voltolina[5]

Received: 21 December 2015 / Accepted: 19 April 2016

Abstract The mercury content of mullets and black mojarras of Urías lagoon (NW Mexico) were determined every second month from November 2012 to September 2013, to determine differences related to season or to trophic levels. The Hg contents of the muscle were significantly higher in mojarras, confirming that Hg contents tend to increase along the food chain, while the levels in liver were higher in mullets, suggesting different Hg storage strategies of these species. In mullets, the content of muscles did not vary seasonally and was significantly lower than in the liver. In black mojarras there were no significant differences between muscle and liver, and the lowest mean values were in May in both tissues. Given the low Hg contents, both species are safe for human consumption, but care should be taken in traditional fishing communities.

Keywords Mercury content · Mullet · Black mojarra · Food habits · Seasonal differences · Coastal lagoons · PTWI

✉ D. Voltolina
voltolin04@cibnor.mx

[1] Facultad de Ciencias del Mar, Universidad Autónoma de Sinaloa, Paseo Claussen s/n, 82000 Mazatlán, Sinaloa, Mexico

[2] Programa de Posgrado en Recursos Acuáticos, Facultad de Ciencias del Mar, Universidad Autónoma de Sinaloa, 82000 Mazatlán, Sinaloa, Mexico

[3] Instituto Tecnológico de Mazatlán, 82000 Mazatlán, Sinaloa, Mexico

[4] Universidad Autónoma de Nayarit, Tepic, Nayarit, Mexico

[5] Laboratorio de Estudios Ambientales UAS-CIBNOR, P.O. Box 1132, 82000 Mazatlán, Sinaloa, Mexico

Mercury (Hg) is a highly toxic pollutant of worldwide concern. It reaches terrestrial and aquatic environments mainly through aerial deposition and accumulates eventually in coastal waters and sediments through river transport and continental runoff. It may be present in several organic and inorganic forms, among which methylated mercury (MeHg) is considered its most toxic species (EFSA 2012, Polak-Juszczak 2015). In Mexico, total atmospheric emissions for 2004 were estimated at 50.46 Mg of Hg, in agreement with the 19.7–97.4 Mg range estimated by UNEP (2013) for Central America, including Mexico and the Caribbean. The main sources were Hg-based paints (23.1 Mg), waste burning (7.36 Mg), cement manufacturing (4.9 Mg), mineries (4.44 Mg), coal burning power plants and chlor-alkali plants, with 2.96 and 2.75 Mg, respectively (Maiz-Larralde 2008). Estimates of releases to the terrestrial environment amount to 185.66 Mg, mostly in mine tailings (155.39 Mg) and informal waste deposits (28.7 Mg), and those to national waters from wastewater treatment systems, dental works residues and Hg-based paints were estimated as 6.13 Mg (Maiz-Larralde 2008).

Estero de Urías lagoon (State of Sinaloa, NW Mexico) is of particular interest because most lagoons of this state are impacted by effluents of intensive agriculture and related activities (Páez-Osuna and Osuna-Martínez 2015), while Urías Lagoon is mostly under urban impact. This is generated by the city of Mazatlán and its commercial and fishing harbor, the related industrial activities (mostly seafood processing plants), one thermoelectric power plant, and unknown volumes of untreated urban wastewater of the surrounding areas. Additional impacts are the effluents of five shrimp farms (Ochoa-Izaguirre and Soto-Jiménez 2014).

Fish is considered important for human nutrition because of its high content of essential nutrients. Its consumption has several dietetic advantages and additionally it decreases the risk of cardiovascular diseases and neuro-development disorders, among others (FDA 2006). However, fish is known to accumulate metals, partly from water and mainly through the food chain and, given the affinity of MeHg for muscle tissues (Renzoni et al. 1998), its consumption may entail an unknown degree of risk. Since Hg excretion rates are generally lower than the rates of inputs, its concentrations tend to increase progressively along the food chain (Sackett et al. 2013). This increases the level of risk if the fish consumed is a predator species.

In this study we determined the Hg content of the muscle and liver of mullets, *Mugil* spp, which are omnivorous and detritivorous, and of the black mojarra *Eugerres*, which is a carnivorous predator and which, for this reason, was expected to have a higher Hg content than mullets. Since both species are popular in fish markets, the data obtained were used to assess the level of risk associated to their consumption and calculate their provisional tolerable weekly intake (PTWI). Season-related differences in the Hg content in aquatic organisms have been described in Mexican NW lagoons, with higher values during the rainy season because of water transport and continental runoff (Páez-Osuna and Osuna-Martínez 2015). However, inputs to Urías lagoon are continuous, which might cause differences in the time-related trends of Hg contents of the two species.

Materials and Methods

Samples were obtained in Estero de Urías every 2 months, from November 2012 to September 2013. On each date, three samples of 10 adult fish of similar size (annual mean: *Mugil* spp = 30.4 ± 3.6 cm; *E. axillaris* = 24.6 ± 3.2 cm) were obtained from local fishermen at their main landing site (106° 25′W, 23° 10′N), and transported to the laboratory on ice in individual polyethylene bags. The 10 specimens of each sample were dissected and equal aliquots of each tissue were used to obtain composite samples. These were freeze-dried, ground, homogenized in a Teflon mortar and three 0.5 g aliquots were pre-digested overnight at room temperature with 5 mL of HNO_3 (trace metal grade). Digestion was carried out in Teflon bombs at 120 °C for 4 h, each digested sample was diluted to 25 mL with Milli-Q water, and total mercury was determined after reduction with $SnCl_2$ in a Buck Scientific model 410 mercury analyzer. For QA/QC all tissues were analyzed in triplicate: detection limit was 0.01 µg/g, the certified reference material DOLT-4 gave 104.34 % recovery. Blanks were used to check contamination and all materials were acid-washed.

Risk due to fish consumption was estimated with the equation HQ = E/RfD (Newman and Unger 2002), where HQ is the hazard quotient, E and RfD are the level of Hg exposure and the reference dose for total Hg (0.5 µg/kg/day for an adult: FDA 2006), respectively, and the level of exposure E is obtained as E = C I/W, where C is Hg concentration (µg/g wet weight) of the food item, I is its apparent daily consumption (1.29 and 0.08 kg/person/year for mojarras and mullets, equivalent to 3.53 and 0.22 g/day, respectively: CONAPESCA 2011). W is the bodyweight (bw) of the average Mexican consumer: 70.7 kg and 62.9 kg for women of child bearing age (CANAIVE 2012).

Risk due to mullet and black mojarra consumption was also estimated using for these two species the total Hg:MeHg ratio 1:0.8 determined for several fish species by Polak-Juszczak (2015).

The mean values of the three samples of muscle and liver of each month were considered representative for the specimens of each species caught in that particular month, and were compared using one way ANOVA or Kruskall-Wallis tests, depending on the results of Kolmogorov–Smirnov and Bartlett's tests. Significantly different means were separated with Holm-Sidak or Dunn's tests. The mean values determined in each sample of muscle of each species were compared to the corresponding samples of livers with t tests for paired observations. Differences between species were determined with t tests. The software for all tests was SigmaStat 3.5 (Systat Software, San Jose, CA), and the level of significance used for all tests was $\alpha = 0.05$.

Results and Discussion

Mullet samples consisted in almost equal parts of *Mugil cephalus* and *M. curema* which, in view of their common habitat and feeding habits (Sánchez-Rueda 2002), were not separated for analysis. There were no significant differences between the mean concentrations determined in the muscle, which ranged from 0.09 ± 0.06 (March) to 0.23 ± 0.08 µg/g (September). In the liver, mean concentrations tended to be higher between March and July, and the highest was that of the May samples (Table 1).

Although Hg contents in aquatic biota are supposed to be higher in the rainy season (Páez-Osuna and Osuna-Martínez 2015), the highest mean value in the liver was found in May, towards the end of the dry season and the lowest in September, at the peak of the rainy season (Table 1). The mean Hg concentration in the muscle of *E. axillaris* was significantly higher than that of *Mugil* spp (0.52 ± 0.36 and 0.16 ± 0.07 µg/g, respectively) whereas that of liver was significantly higher in *Mugil* spp (0.61 ± 0.30 and 0.83 ± 0.32 µg/g for *E. axillaris* and *Mugil* spp). The liver of mullets had significantly higher values than the muscle (t test for paired samples, $p < 0.05$),

Table 1 Monthly mean values (±standard deviation) of the total mercury content (μg/g, dw) of muscle and liver of *Mugil* spp of Estero de Urías lagoon

Month	*Mugil* sp	*E. axillaris*
Muscle		
Nov	0.18 ± 0.04a	0.70 ± 0.24b
Jan	0.18 ± 0.06a	0.62 ± 0.32b
Mar	0.09 ± 0.06a	0.53 ± 0.16b
May	0.12 ± 0.06a	0.10 ± 0.01a
Jul	0.17 ± 0.05a	0.55 ± 0.09b
Sep	0.23 ± 0.08a	0.63 ± 0.15b
Liver		
Nov	0.66 ± 0.16ab	0.86 ± 0.08b
Jan	0.79 ± 0.09ab	0.70 ± 0.23ab
Mar	1.02 ± 0.11ab	0.26 ± 0.12a
May	1.17 ± 0.32b	0.29 ± 0.30ab
Jul	0.96 ± 0.24ab	0.75 ± 0.16ab
Sep	0.36 ± 0.06a	0.80 ± 0.15ab

Different letters indicate significant differences between data in the same column (one way ANOVAs, $\alpha = 0.05$)

while the difference between tissues was not significant for *E. axillaris* ($p > 0.05$).

Although Delgado-Alvarez et al. (2015) did not find any season-related differences in the Hg contents (0.05–0.37 μg/g) of oysters of the NW coast of Mexico, several authors (Yesudhason et al. 2013; Páez-Osuna and Osuna-Martínez 2015) reported a tendency to higher Hg contents in aquatic organisms during the rainy season, probably related to increased inputs due to surface runoff from the surrounding agricultural areas. The different trends of fishes of Urías lagoon are probably due to the continuous inputs of urban wastewater, industrial effluents and fumes and cooling water from an oil-burning power plant (Ochoa-Izaguirre and Soto-Jiménez 2014).

The values determined in the muscles of *Mugil* spp in this study are comparable to those found in other Mugilidae of NW Mexico, but higher than those of *M. incilis* of the Colombian coast, even in Cartagena Bay, where sediments are contaminated by the residues of a chlor-alkali plant (Alonso et al. 2000). Additionally, our data coincide with most authors, which indicate higher concentrations in the liver than in the muscle (Table 2).

Since Mugilidae are mostly primary consumers or detritus feeders, their Hg contents were expected to be lower than in *E. axillaris* (troph 2–2.5 and 3.4, respectively: Froese and Pauly 2014). As expected, the mean Hg content of the muscle of *E. axillaris* was significantly higher than that of *Mugil* spp, although this had significantly higher values in the liver. This, the lack of a significant difference between tissues of *E. axillaris*, and the significantly higher value in the liver of *Mugil* spp, indicate that these two species accumulate Hg in different tissues.

The values found in *E. axillaris* of Estero de Urías lagoon coincide with the range of mean values determined in Latin American samples of the same taxonomic group (Gerreidae) (Table 2). Liver is the preferential organ for metal accumulation because of its physiological functions of detoxification and storage (Tepe et al. 2008), which coincides with the difference between tissues determined in *Mugil* spp, but not with the results obtained with *E. axillaris* which shows that, at least for Hg, metal accumulation in the liver is not a general rule.

On the other hand, due to the presence of sulfhydril groups, methylated Hg (MeHg) is supposed to have higher affinity for the muscle (Renzoni et al. 1998). This would explain the similar values found in the liver and muscle of *E. axillaris*, and coincides with the lack of difference between hepatic and muscular tissues and/or the higher Hg concentrations of the muscles determined in several freshwater and marine fish species by Cizdziel et al. (2003) and Polak-Juszczak (2015), among others.

However, in agreement with our results for *Mugil* spp, other authors determined higher Hg values in fish liver than in muscle (i.e. Azevedo et al. 2012), which might be due to differences in the Hg species present in these two organs, since the low values in the muscle with corresponding high liver contents might be explained by a high percentage of inorganic Hg, probably because of ongoing demethylation of MeHg in the liver (Joiris et al. 1999), whereas similar or higher values in the muscles seem to indicate a higher percentage of total Hg present as MeHg (Branco et al. 2007, Coelho et al. 2010). This could be due to different bioaccumulation and detoxification strategies (Sackett et al. 2013), leading to interspecific differences such as those found in this study.

Present Mexican regulations indicate that the maximum Hg content (as MeHg) in marine products other than tuna and related species should not exceed 0.5 μg/g (wet weight) (Secretaría de Salud 2011), while the European Community established a maximum total Hg limit of 1.0 μg/g (wet weight) for most fish (European Union 2008). The values detected in our study, converted to wet weight assuming moisture content of 80 % of fresh fish, and a total Hg: MeHg ratio of 1:0.8, are well below the maximum permissible contents of both limits. The mean HQ values calculated for mullet consumption were $0.25 \pm 0.08 \times 10^{-3}$ and $1.26 \pm 0.38 \times 10^{-3}$ for total Hg and MeHg, respectively, which were significantly ($p < 0.01$) lower than those for black mojarra ($1.29 \pm 0.53 \times 10^{-2}$ and 0.06 ± 0.03. These low values seem to indicate that in Mexico there is a negligible risk related to the consumption of these two fish species.

Table 2 Mercury contents (µg/g dw) in *Mugil* spp., and in *Eugerres* and other Gerreidae from Mexico and other Latin American countries

Species	Zone	Muscle	Liver	Troph*
Mugil spp[a]	Urías lagoon, Mexico	0.09–0.23	0.36–1.17	2.0–2.5
Mugil cephalus[b]	Coastal lagoons NW Mexico	0.02–0.86	0.26–1.33	2.5 ± 0.2
Mugil cephalus[c]	Topolobampo, NW Mexico	0.05	1.22	2.5 ± 0.2
Mugil incilis[d]	Cartagena Bay, Colombia	0.01–0.09	–	2.0 ± 0.1
Mugil incilis[d]	Ciénaga Santa Marta, Colombia	0.01–0.02		2.0 ± 0.1
Eugerres axillaris[a]	Urías lagoon, Mexico	0.10–0.70	0.26–0.86	3.4 ± 0.5
Eugerres plumieri[d]	Cartagena Bay, Colombia	0.11–0.33	–	2.2 ± 0.0
Eugerres plumieri[d]	Ciénaga Santa Marta, Colombia	0.01–0.03	–	2.2 ± 0.0
Eugerres brasilianus[e]	Jaguaribe River, NE Brazil	0.02–0.13		3.4 ± 0.4
Diapterus peruvianus[c]	Topolobampo, NW Mexico	0.5	–	3.7 ± 0.2
Eugerres plumieri[f]	Puerto Rico	0.05–0.21	–	2.2 ± 0.0

* Trophic level, based on food ítems (Froese and Pauly 2014)

[a] This study; [b] Ruelas-Inzunza and Páez-Osuna (2005); [c] Ruelas-Inzunza et al. (2008); [d] Alonso et al. (2000); [e] Costa and Lacerda (2014); [f] Rodríguez-Sierra and Jiménez (2002)

However, the risk assessed using the only available values of fish and seafood consumption (apparent consumption, calculated considering total national landings and the total Mexican population) might give an optimistic estimate of the risk of Hg ingestion. A recent survey in the coastal city of Mazatlán (Delgado-Alvarez 2015) evaluated a higher local consumption than the national value. While all HQs remained below the level of risk even considering local consumption rather than the national value, the coefficients were at least twice higher than those estimated using official data, which indicates the urgent need of further studies to identify eventual populations at risk, which might be some of the traditional fishing communities common along the Mexican Pacific coast.

The European Community suggests a provisional tolerable weekly intake (PTWI) of 4 µg/Kg bw of total Hg (including pregnant women), but the limit for MeHg is only 1.3 µg (EFSA 2012), which is lower than the 1.6 µg established by FAO-WHO (2003). Using the lower PTWI value (EFSA 2012), the tolerable weekly consumption would be 2.84 kg/person for mullet and 0.87 kg/person for black mojarra for an average adult (2.54 and 0.78 kg in the case of women of child-bearing age, with an average bw of 62.9 kg), which become likely quantities in the fishermen's families of the poorest Mexican coastal communities.

Acknowledgments Supported by projects PROFAPI-UAS 2014/074; PROMEP/103.5/13/9354 and CONACYT INFRA 2012-01-188029. Karla Sánchez helped with laboratory work, and I.P. Rosas with cabinet and editing work.

References

Alonso D, Pineda P, Olivero J, González HH, Campos N (2000) Mercury levels in muscle of two fish species and sediments from the Cartagena Bay and the Ciénaga Grande de Santa Marta, Colombia. Environ Pollut 109:157–163

Azevedo JS, de Souza-Sarkis JE, Oliveira TA, Ulrich T (2012) Tissue-specific mercury concentrations in two catfish species from the Brazilian coast. Braz J Oceanogr 60:211–219

Branco V, Vale C, Canário J, Neves dos Santos M (2007) Mercury and selenium in blue shark (*Prionace glauca*, L. 1758) and swordfish (*Xiphias gladius*, L. 1758) from two areas of the Atlantic Ocean. Environ Pollut 150:373–380

CANAIVE (2012) ¿Cuánto mide México?. Cámara Nacional de la Industria del Vestido, Mexico

Cizdziel J, Hinners T, Cross C, Pollard J (2003) Distribution of mercury in the tissues of five species of freshwater fish from Lake Mead, USA. J Environ Monit 5:802–807

Coelho JP, Santos H, Reis AT, Falcão J, Rodrigues ET, Pereira ME, Duarte AC, Pardal MA (2010) Mercury bioaccumulation in the spotted dogfish (*Scyliorhinus canicula*) from the Atlantic Ocean. Mar Pollut Bull 60:1372–1375

CONAPESCA (2011) Anuario estadístico de acuacultura y pesca. Comisión Nacional de Pesca, Mazatlán

Costa BGB, Lacerda LD (2014) Mercury (Hg) in fish consumed by the local population of the Jaguaribe River lower basin, Northeast Brazil. Environ Sci Pollut Res 21:13335–13341

Delgado-Alvarez CG (2015) Mercurio total en recursos pesqueros de importancia comercial en el Noroeste de México: riesgo potencial a la salud humana. Ph. D. Dissertation, Universidad Autónoma de Sinaloa

Delgado-Alvarez CG, Ruelas-Inzunza J, Osuna-López JI, Voltolina D, Frías-Espericueta MG (2015) Total mercury content in cultured oysters from NW Mexico: health risk assessment. Bull Environ Contam Toxicol 94:209–213

EFSA (2012) Panel on contaminants in the food chain (CONTAM); Scientific opinion on the risk for public health related to the presence of mercury and methylmercury in food. EFSA J 10:2985. p 241. www.efsa.europa.eu/efsajournal. Accessed 14 Dec 2015

European Union (2008) Commission regulation (ec) no 629/2008 of 2 July 2008 amending regulation (ec) no 1881/2006 setting maximum levels for certain contaminants in foodstuffs. Official Journal of the European Union L 173/6, 3.7.2008

FAO-WHO (2003) Summary and conclusions of the sixty-first meeting of the Joint FAO/WHO Expert Committee on Food Additives (JECFA), Annex 4. JECFA/61/SC. ftp://fao.org/es/esn/jecfa/jecfa61sc.pdf. Accessed 19 Dec 2015

FDA (2006) Mercury levels in commercial fish and shellfish. Washington DC. www.cfsan.fda.gov/~frf/sea-mehg.html Accessed 1 Sep 2015

Froese R, Pauly D (2014) FishBase. World Wide Web electronic publication. http://www.fishbase.org/. Accessed 1 May 2015

Joiris CR, Holsbeek L, Moatemri NL (1999) Total and methylmercury in sardines *Sardinella aurita* and *Sardina pilchardus* from Tunisia. Mar Pollut Bull 38:188–192

Maiz-Larralde P (2008) Final-Inventario Nacional de Liberaciones de Mercurio-México 2004. Final report. Centro Nacional de Investigación y Capacitación Ambiental, Instituto Nacional de Ecología, SEMARNAT

Newman MC, Unger MA (2002) Fundamentals of ecotoxicology. Lewis Publishers, Boca Raton

Ochoa-Izaguirre MJ, Soto-Jiménez MF (2014) Variability in nitrogen stable isotope ratios of macroalgae: consequences for the identification of nitrogen source. J Phycol 51:46–65

Páez-Osuna F, Osuna-Martínez C (2015) Bioavailability of cadmium, copper, mercury, lead, and zinc in subtropical coastal lagoons from the southeast Gulf of California using mangrove oysters (*Crassostrea corteziensis* and *Crassostrea palmula*). Arch Environ Contam Toxicol 68:305–316

Polak-Juszczak L (2015) Selenium and mercury molar ratios in commercial fish from the Baltic Sea: additional risk assessment criterion for mercury exposure. Food Control 50:881–888

Renzoni A, Zino F, Franchi E (1998) Mercury levels along the food chain and risk for exposed populations. Environ Res 77A:68–72

Rodriguez-Sierra CJ, Jiménez B (2002) Trace metals in striped mojarra fish (*Diapterus plumieri*) from Puerto Rico. Mar Pollut Bull 44:1039–1045

Ruelas-Inzunza J, Páez-Osuna F (2005) Mercury in fish and shark tissues from two coastal lagoons in the Gulf of California, México. Bull Environ Contam Toxicol 4:294–300

Ruelas-Inzunza J, Meza-López G, Páez-Osuna F (2008) Mercury in fish that are of dietary importance from the coasts of Sinaloa (SE Gulf of California). J Food Compos Anal 21:211–218

Sackett DK, Cope WG, Rice JA, Aday DD (2013) The influence of fish length on tissue mercury dynamics: implications for natural resources management and human health risk. Int J Res Public Health 10:638–659

Sánchez-Rueda P (2002) Stomach content analysis of *Mugil cephalus* and *Mugil curema* (Mugiliformes: Mugilidae) with emphasis on diatoms in the Tamiahua lagoon, México. Rev Biol Trop 50:245–252

Secretaría de Salud (2011) Norma Oficial Mexicana NOM-242-SSA1-2009, Productos y servicios. Productos de la pesca frescos, refrigerados, congelados y procesados. Especificaciones sanitarias y métodos de prueba. Diario Oficial, Feb 10 2011

Tepe Y, Türkmen M, Türkmen A (2008) Assessment of heavy metals in two commercial fish species of four Turkish seas. Environ Monit Asses 146:277–284

UNEP (2013) Global mercury assessment 2013: sources, emissions, releases and environmental transport. UNEP Chemicals Branch, Geneva

Yesudhason P, Al-Busaidi M, Al-Rahabi W, Al-Waili AS, Al-Nakhaili AK, Al-Mazrooei NA, Al-Habsi SH (2013) Distribution patterns of toxic metals in the marine oyster *Saccostrea cucullata* from the Arabian Sea in Oman: spatial, temporal, and size variations. SpringerPlus 2:1–11