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Total Mercury Content in Cultured Oysters from NW Mexico: Health Risk Assessment

C. G. Delgado-Álvarez · J. Ruelas-Inzunza · J. I. Osuna-López · D. Voltolina · M. G. Frías-Espericueta

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Abstract The total mercury (Hg) content of the soft tissues of cultured oysters of the genus *Crassostrea* obtained during the dry and rainy seasons at sampling sites of NW Mexico with different degrees of urbanization, was determined by cold vapor atomic absorption spectrophotometry. Hg levels ranged from 0.05 to 0.37 μg/g (dry weight) and no significant differences ($p > 0.05$) related to season and sampling site were observed. The values did not exceed the limit of 1.0 μg/g (wet weight) established by Mexican legislation and by the Food and Drug Agency (FDA), and the hazard quotient was between 0.001 and 0.002. The estimated hazard quotient for MeHg ranged approximately from 0.002 to 0.01.

Keywords Mercury · Oyster farms · NW Mexico · Risk assessment

Mercury (Hg) enters the environment through volcanism, fossil fuel and waste burning and from other human as well as natural processes. It has a high environmental cost due to its non-biodegradable nature and long biological half-life (Hylander and Goodsite 2006). It is a cause of ecological concern, because it accumulates in sediments and in aquatic organisms and is biomagnified along aquatic food chains (Pan and Wang 2011; Jara-Marini et al. 2012; Kim et al. 2012). Among the inorganic and organic Hg species of different toxicity, methylated Hg (MeHg) is well known for its neurotoxicity and because it affects several physiological processes (Ipolyi et al. 2004; Apeti et al. 2012). Although it may originate from human sources, methylation by bacterial activities is its most common and important source.

Oysters of the genus *Crassostrea* have been used as biomonitors of contamination of the aquatic environment, because of their adaptability to environmental conditions, tolerance to contamination and wide geographic distribution (Vaisman et al. 2005). This genus is represented along the Pacific Mexican coasts by four species. Three of these (*C. corteziensis, C. iridescens, C. palmula*) are native, and one (*C. gigas*) was introduced for aquaculture (Rangel-Dávalos 1990). Among these, *C. corteziensis* and *C. gigas* are cultured for human consumption along the NW coast of Mexico, although most sections of the coastline are impacted by the discharges of urban, industrial, mining and agriculture activities.

In this study we determined the concentration of total mercury in oysters of this genus obtained at different sampling sites of three coastal states of NW Mexico during the dry and the rainy season, and used this information to assess human health risk, considering the average rate of consumption in Mexico.

Methods and Materials

Samples were obtained in August and November 2010 (rainy season) and February and May 2011 (dry season)

C. G. Delgado-Álvarez
Aquatic Resources Postgraduate Program, Faculty of Marine Sciences, Autonomous University of Sinaloa, Mazatlán, Sinaloa, Mexico

J. Ruelas-Inzunza
Technological Institute of Mazatlán, Mazatlán, Sinaloa, Mexico

J. I. Osuna-López · M. G. Frías-Espericueta (✉)
Faculty of Marine Sciences, Autonomous University of Sinaloa, Paseo Claussen s/n, 82000 Mazatlán, Sinaloa, Mexico
e-mail: friasm@ola.icmyl.unam.mx

D. Voltolina
Environmental Studies Laboratory, UAS-CIBNOR, Mazatlán, Sinaloa, Mexico

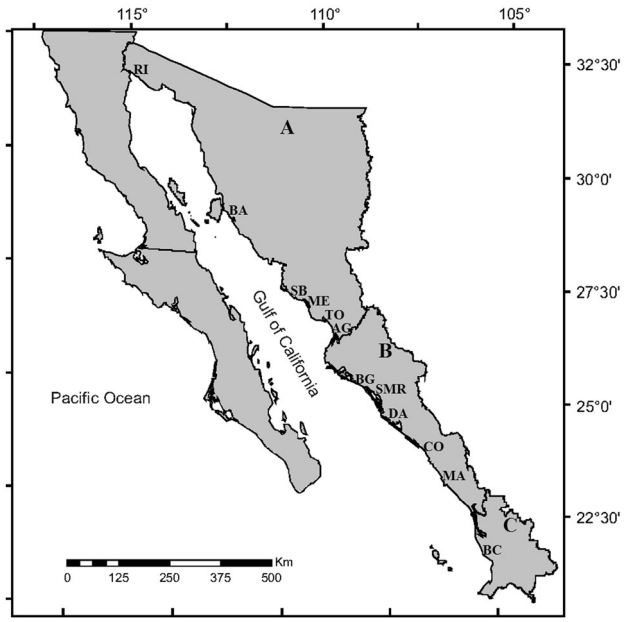

Fig. 1 Locations where oysters were collected. *A*: Sonora State, *B*: Sinaloa State, *C*: Nayarit State. *RI* Estación Riíto, *BA* Bachoco, *SB* Santa Bárbara, *ME* Mélagos, *TO* Tóbari, *AG* Agiabampo, *BG* Brecha Guasave, *SMR* Santa María la Reforma, *DA* Dautillos, *CO* Cospita, *MA* Mármol, *BC* Boca Camichín

from twelve sites (two visits/season in each station) located on the coastal area of NW Mexico (Fig. 1). All oysters (20/sample) were of commercial size (shell height >5 cm) and those of each sample had similar height. After collection, oysters were washed with seawater, transported within 24 h to the laboratory in clean polyethylene bags kept in coolers, and stored at −20°C until processing for metal analysis (Vaisman et al. 2005). Samples from Sonora State were cultured Pacific oyster *C. gigas* (mean size: 9.5 ± 1.6 cm), those from Sinaloa and Nayarit States were pleasure oysters *C. corteziensis* cultured with different techniques (clutchless oysters in Sinaloa, with mean size: 8.9 ± 1.4 cm; traditional culture on mother shell collectors in Nayarit: mean size: 6.8 ± 1.2 cm). Those obtained at Mármol station were wild *C. iridescens* (rock oyster, mean size: 9.5 ± 1.1 cm), collected from their natural habitat.

After thawing, oysters were shucked with a stainless steel knife, the whole soft bodies of the 20 specimens of each sample were freeze dried (72 h at 80 × 10^{-3}mbar, −50°C), ground and homogenized in a Teflon mortar to obtain a composite sample and 0.25 g of the homogenate were pre-digested overnight at room temperature with 5 mL of concentrated nitric acid (Baker, trace metal grade) (Rojas de Astudillo et al. 2002). Digestion was performed in closed Teflon bombs at 120°C for 3 h, and samples were diluted to 25 mL with MilliQ water. All materials and glassware used in sampling and metal analysis were acid cleaned (Moody and Lindstrom 1977).

Total Hg was determined by cold vapor atomic absorption spectrophotometry (Buck Scientific) after reduction with $SnCl_2$ in a mercury analyzer (Buck Scientific) (Ruelas-Inzunza et al. 2004). For QA/QC all samples were analyzed in triplicate, the accuracy of the analytical method was evaluated with certified reference material (DORM 3) with a mean recovery of 104.3 % ± 1.0 % and parallel reagent blanks were used to check contamination. The limit of detection was 0.003 μg/g.

The values obtained in the dry and in the rainy season at each sampling station were compared with a two ways ANOVA test with $\alpha = 0.05$ using site and sampling season as independent variables. The test was run after rank transformation, because the data were not homoscedastic (Conover and Iman 1981; Zar 1999; Conover 2012). The risk to human health due to oyster consumption was estimated according to Newman and Unger (2002), using the equation HQ = E/RfD, in which 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: FDA 2006), respectively, and the level of exposure E is calculated as E = C I/W where C is the total concentration of Hg (in μg/g wet weight) of the species tested, I is its apparent daily consumption (0.39 kg/person/year = 1.07 g/person/day: CONAPESCA 2011). W is the assumed weight of an adult consumer (70 kg).

The risk due to MeHg content of the oysters was estimated with the same equations, using 0.1 as RfD (EPA 2001) and the average MeHg/total Hg ratio calculated using the data available for oysters in Claisse et al. (2001), Pan and Wang (2011), and Apeti et al. (2012).

Results and Discussion

Hg values ranged from 0.05 to 0.34 μg/g (dry weight), determined during the rainy season at Santa Barbara and Tóbari, respectively. This range is comparable to that found by Apeti et al. (2012) in the Northern Gulf of Mexico. There were no significant differences due to site or sampling season. The annual mean values ranged between 0.12 and 0.27 μg/g (station Dautillos, Sinaloa and station Bachoco, Sonora, respectively), and the overall mean concentration, calculated using all the data obtained at each station was 0.172 ± 0.034 μg/g (Table 1).

This lack of difference may be explained by the Hg sources shared by the three states: although the production of the mining industry of Sonora is far higher than those of Sinaloa and Nayarit, the freshwater inputs to the coastal waters of these states flow from the mineral-rich Sierra

Table 1 Range, annual mean total Hg content (±SD) of *Crassostrea* spp. from different farms in NW Mexico, hazard quotient (HQ), and estimated MeHg content and respective hazard quotient

Site	Range	(Hg) μg/g DW	HQ THg	(MeHg)[b] μg/g DW	HQ MeHg[b]
RI	0.093–0.292	0.138 ± 0.049[a]	0.0010	0.054	0.0020
BA	0.166–0.186	0.175 ± 0.009[a]	0.0019	0.099	0.0097
SB	0.150–0.374	0.254 ± 0.093[a]	0.0013	0.068	0.0067
ME	0.161–0.201	0.175 ± 0.022[a]	0.0013	0.068	0.0067
TO	0.092–0.167	0.139 ± 0.041[a]	0.0014	0.072	0.0070
AG	0.129–0.264	0.177 ± 0.063[a]	0.0013	0.069	0.0068
BG	0.113–0.169	0.140 ± 0.025[a]	0.0010	0.055	0.0053
SMR	0.152–0.241	0.189 ± 0.045[a]	0.0014	0.074	0.0073
DA	0.005–0.195	0.119 ± 0.080[a]	0.0009	0.047	0.0045
CO	0.135–0.173	0.157 ± 0.016[a]	0.0012	0.061	0.0060
MA	0.113–0.200	0.166 ± 0.039[a]	0.0012	0.065	0.0063
BC	0.106–0.261	0.193 ± 0.066[a]	0.0014	0.075	0.0074
Mean	–	0.172 ± 0.034	0.0013	0.068	0.0063

The equal letters indicate lack of significant differences (two ways ANOVA, $\alpha = 0.05$)

[b] Calculated using 0.3915 as mean MeHg/total Hg ratio, estimated from the ranges reported for oysters by Claisse et al. (2001), Pan and Wang (2011), and Apeti et al. (2012). Total range of MeHg/total Hg ratios 0.156–0.621

Table 2 Range of mean Hg concentrations (μg/g, dry weight) in oysters of different areas of the world

Species	Area	Hg	References
Crassostrea corteziensis	Bacochibampo Bay, Sonora State, Mexico	0.12–0.16	García-Rico et al. (2010)
Crassostrea gigas	Coastal lagoons, Sinaloa State, Mexico	0.06–0.91	Osuna-Martínez et al. (2010)
Crassostrea corteziensis	Coastal lagoons, Sinaloa State, Mexico	0.16–0.58	Osuna-Martínez et al. (2010)
Crassostrea corteziensis	Urías lagoon, Sinaloa State, Mexico	0.03–0.08	Jara-Marini et al. (2012)
Crassostrea virginica	Campeche, Gulf of Mexico, Mexico	0.40–2.00	Aguilar et al. (2012)
Crassostrea virginica	Northern Gulf of Mexico, USA	0.03–0.5	Apeti et al. (2012)
Crassostrea rhizophorae	Villa Clara, Cuba	0.19–0.69	Olivares-Rieumont et al. (2012)
Crassostrea gigas	Minamata Bay, Japan	10	Eisler (1987)
Crassostrea sp.	Gulf of Paria, Sucre State, Venezuela	0.08–0.68	Rojas de Astudillo et al. (2002)
Saccostrea cucullata	Zhejiang, China	0.11 ± 0.04	Fang et al. (2004)
Saccostrea cucullata	Qeshm Island, Iran	0.03–0.06	Shirneshan et al. (2012)
Crassostrea rhizophorae	Northeast Brazil	0.02–0.30	Vaisman et al. (2005)
Crassostrea gigas	Ebro Delta, Spain	0.12–0.27	Ochoa et al. (2013)
Ostrea edulis	Coast of Croatia	0.024–2.72	Bogdanović et al. (2014)
Saccostrea cucullata	Arabian Sea, Oman	0.07 ± 0.03	Yesudhason et al. (2013)
Crassostrea spp.	NW coast, Mexico	0.12–0.25	This study

Madre Occidental (states of Chihuahua, Durango and Zacatecas) where >70,000 ton of Hg are still present in mine tailings as residues of the >190,000 tons of Hg used from colonial times until the early 1900s in the amalgamation process involved in silver and gold extraction (Castro-Díaz 2013).

Important additional sources are the effluents from the 3.5×10^6 ha of fertile Pacific coastal plains dedicated to intensive agriculture, since Hg-based fungicides are still used for pest control of several crops (Osuna-Martínez et al. 2010). Other inputs are atmospheric deposition, because of Hg volatility which allows easy transport even to areas far from the geographic source (Vaisman et al. 2005; Apeti et al. 2012), and the Hg content of seawater, because of the continuous water renovation of the coastal lagoons and embayments (Osuna-Martínez et al. 2010; Harris et al. 2012).

There were no differences between the values determined in the rainy and dry season at each sampling site, but values tended to be higher in the rainy season. This might

explain the higher metal levels (including Hg) found in oysters during the rainy than the dry season by Osuna-Martínez et al. (2010) in some coastal lagoons of Sinaloa State, who related this difference to increased continental runoff and concurrent high terrigenous sediment transport from metal-rich watersheds because of natural processes, such as rock weathering and leaching, or from metal-impacted surrounding areas (Yesudhason et al. 2013).

A comparison of the Hg levels in the soft tissues of oysters from several parts of the world show that, apart from the extremely high values reported for Minamata Bay, the range of data found in this study (0.05–0.37 μg/g) lies within the limits of most values found in literature, with the exception of those reported in Zhejiang, China and in the transition zone between the Persian Gulf and Arabian Sea (Oman and Qeshm Island, Iran), although with a tendency to be lower than the values reported for Villa Clara, Cuba and for the coast of Croatia. As to previous studies in NW Mexico, the range found by García-Rico et al. (2010) was similar to that of this study, the upper limits of those obtained by Osuna-Martínez et al. (2010) for *C. gigas* and *C. corteziensis* were between twice and three times higher, whereas the upper limit of the Hg content of the wild oysters of Urías lagoon (Jara-Marini et al. 2012) was lower than the lowest of our mean values (Table 2).

Due to its high toxicity even at low concentrations, to the higher toxicity and high affinity to biological systems of the end products of its biotransformation, especially of methyl-mercury (Clarkson and Magos 2006), risk assessments of Hg exposure should be carried out in all worlds coastal zones. For instance, Shirneshan et al. (2012) reported no risk by human consumption in view of the low values detected in Qeshm Island (Persian Gulf), and a similar result was obtained by Bogdanović et al. (2014) in their biomonitoring survey along the coast of Croatia. In the case of this study, all HQ values were <1 (Table 1) indicating no risk for oyster consumption in NW Mexico.

In general, because of their low trophic position, bivalves have lower Hg content than fishes (Jara-Marini et al. 2012; Kim et al. 2012). However their contents may reach toxic levels for top predators because of biomagnification through the food web, which is of ecological as well as human health concern (Fang et al. 2004; Ochoa et al. 2013). The oysters used for this study did not exceed the 1.0 μg/g (wet weight) limit established by Mexican legislation (SSA 1995) and FDA (2006) and the calculated HQ was <1. These values indicate that at least for the time being the Hg concentration of cultivated oysters is not a matter of concern although, in view of increasing urbanization and industrialization of NW Mexico, the Hg content of aquatic organisms should be monitored in order to prevent human health risk.

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