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Mercury in raw and cooked shrimp and mussels and dietary Brazilian exposure

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ABSTRACT

Seafood is the main source of dietary mercury (Hg) exposure and it can be affected by species, origin, tissue, and processing. The objective of this study was to investigate total Hg in shrimp and mussels from the retail market, compare species, body tissue distribution and cooking time, and to estimate dietary exposure. A total of 126 samples (whiteleg and Atlantic seabob shrimp and South American mussel) were analyzed. Total mercury was quantified by combustion atomic absorption spectrometry with gold amalgamation. Every sample had total Hg lower than maximum legislation levels. Higher mean levels were found in mussels compared to shrimp; and whiteleg shrimp had higher mean Hg levels compared to Atlantic seabob ($p < 0.05$). Mercury levels in the muscle were higher than in the exoskeleton. Boiling for up to 4 and 6 min did not affect Hg levels in shrimp and mussels, respectively. The estimated methylmercury (MeHg) mean intake from the consumption of shrimp and mussels (0.04–0.09 μg/kg bw for 3 portions/week consumption) was low, less than 6% of the provisional tolerable weekly intake – PTWI.

1. Introduction

In the last decades, the world per capita consumption of seafood has practically doubled. The worldwide production of seafood in 2016 was 17.1 million tons of mollusks and 7.9 million tons of crustaceans, representing 21.4% and 9.8% of all aquaculture, respectively (FAO, 2018). In fact, shellfish are valued today as they add resilience to the global food system (Suplicy, 2020). A diet high in fish and shellfish is beneficial to health (Cobb & Ernst, 2006; Lund, 2013). Seafood, including shrimp and mussels, are rich in omega-3 polyunsaturated fatty acids which are associated with a reduced risk of cardiovascular disease, stroke, and diabetes (Smith & Guentzel, 2010; Vilavert, Borrel, Nadal, Jacobs, & Domingo, 2017). They also have a well-balanced amino acid composition and provide significant levels of a number of other potentially protective components, including vitamins D and B12, selenium, iodine, and choline (Cobb & Ernst, 2006; Smith & Guentzel, 2010; Lund, 2013; Maulvault, Cardoso, Nunes, & Marques, 2013; Cantoral, Batis, & Basu, 2017; Jacobs, Sioen, Jacxsens, Domingo, & Verbeke, 2017; Ralston, Kaneko, & Raymond, 2019). In addition, mussels are sources of taurine, associated with neuronal development (Lund, 2013), and they can lower the incidence of rheumatoid arthritis (Cobb & Ernst, 2006; Suplicy, 2020). However, these protective effects may be counteracted by the presence of mercury (Smith & Guentzel, 2010).

Fish are widely investigated for total mercury and methylmercury; the latter being the more toxic form; however, scarce information is available regarding mercury in shrimp and mussels. The benthic and detritivorous characteristic of shrimp makes it a possible mercury bioaccumulator and a biomarker of environmental contamination (Kaya & Turkoglu, 2017). This is also true for mussels due to their feeding characteristics as filters (Belabed et al., 2013; Ansari et al., 2016; Diop & Amara, 2016; Belivermiş et al., 2019).

In contrast to the mercury risk, shrimp and mussels can be good

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Abbreviations: BW, body weight; DWB, dry weight basis; LOQ, limit of quantification; MeHg, methylmercury; PTWI, provisional tolerable weekly intake.

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sources of selenium (Kehrig, Seixas, Dibenedito, & Malm, 2013; Silva, Viana, Onofre, Korn, & Santos, 2016). In fact, besides showing strong antioxidant activity, selenium is required for the activities of selenoenzymes with critical roles in fetal brain development, growth and thyroid hormone metabolism (Lund, 2013; Monastero, Karimi, Silbernagel, & Meliker, 2016; Ralston & Raymond, 2018). Methylmercury (MeHg) toxicity primarily occurs through disruption of brain selenium metabolism, which can be prevented in an excess of selenium (Se:Hg molar ratio above 1) (Bjørklund et al., 2017; Ralston & Raymond, 2018; Spiller, 2018).

Whiteleg shrimp (*Litopenaeus vannamei*), Atlantic seabob shrimp (*Xiphopenaues kroyeri*) and the bivalve mollusk *Perna perna* – South American rock mussel (Mollusca: bivalvia) are important commercial shellfish in Brazil (MPA, 2012). Whiteleg shrimp is the major specie produced in aquaculture worldwide (FAO, 2018), and its production in Brazil is concentrated mainly in the Northeastern coast (Kubitza, 2015). Atlantic seabob shrimp has wide distribution throughout the Brazilian coast (Costa, Fransozo, Freire, & Castilho, 2007), but mainly captured in the South and Southeast (MPA, 2012). South American rock mussel is the main marine mollusk captured and cultivated in the Southern and Southeastern Brazilian coast (MPA, 2012).

The objective of this study was to determine total mercury in commercial shrimp and mussels and also to investigate the influence of some factors (species, tissue and heat treatment) on mercury level in the studied seafood. The data gathered was used to estimate the exposure of the Brazilian population to methylmercury present in shrimp and mussels using the deterministic model. In addition, the theoretical protective effect of selenium from literature data on mercury toxicity was calculated.

2. Material and methods

2.1. Experimental design

Samples of frozen whiteleg shrimp (*Litopenaeus vannamei*), Atlantic seabob shrimp (*Xiphopenaeus kroyeri*) and South American rock mussel (*Perna perna*) were purchased at the retail market of Belo Horizonte, MG, Brazil from June to December 2017. The samples were transported in isothermal boxes containing crushed ice and were taken immediately to the laboratory.

For the analysis of total mercury content, the shrimp samples were peeled and cleaned (removal of the cephalothorax) and the mussel samples were shelled. Exoskeletons of selected samples of whiteleg shrimp were kept for mercury analysis separately. All samples were minced, homogenized, and stored individually at −18 °C. A total of 126 samples were analyzed, in triplicate, with 42 samples of each species (whiteleg shrimp, Atlantic seabob shrimp and South American rock mussel).

The influence of heat treatment on the contents of total mercury in the whiteleg shrimp and mussels was investigated using three different pools for each cooking time. The boiling time was established according to culinary practices: 2 and 4 min for shrimp and 3 and 6 min for mussels. The moisture content was determined for these pools by drying samples in air circulating oven at 105 °C for 24 h (AOAC, 2016).

2.2 Mercury analysis

The total mercury content of the samples was determined using combustion atomic absorption spectrometry with gold amalgamation (Soares et al., 2018) using a Direct Mercury Analyzer® (DMA-80, Milestone, Sorisole, Italy). A 1000 ± 2 mg/L standard solution of inorganic mercury (Sigma-Aldrich Co., St. Louis, MO, USA) was used to prepare solutions of different concentrations (1.0–8.5 ng of mercury, six equally spaced points) using high-purity water with resistivity of 18.2 MΩ cm (Milli-Q Plus, Millipore, Bedford, MA, USA) and quartz boats for construction of the analytical curves ($R^2 \geq 0.9969$). Total mercury concentrations were obtained by interpolation of absorbance readings at 253.7 nm in external analytical curves. The lower point of the calibration curve was considered the limit of quantification (LOQ), confirmed by six independent replicates (EC, 2002).

Samples (100 mg) were weighed directly into nickel boats and mercury was quantified in the DM Analyzer®. For quality assurance, periodically, empty nickel boats were analyzed to confirm that no residue of mercury was carried over between samples. Blank readings were typically <0.01 absorbance units, corresponding to <0.01 ng of Hg.

2.3. Estimation of Brazilian exposure to methyl mercury by shrimp and mussel consumption and potential theoretical protective effect from selenium

Chronic exposure of the Brazilian population to methylmercury present in seafood was calculated by the deterministic model (IOMC, 2008), using the equation: Exposure = [concentration of methylmercury (μg/kg) x food consumption (kg)]/body weight (kg). Methylmercury occurrence was estimated from total mercury values assuming that 100% mercury was in the methylmercury form in shrimps and 90% in mussels considering mercury speciation data (Batista, Rodrigues, Souza, Souza, & Barbosa Jr, 2011). Data on the Brazilian consumption of shrimp and bivalve mollusks is available as microdata from a dietary survey conducted by the Brazilian Institute of Geography and Statistics (IBGE, 2011), between 2008 and 2009 for a population over 10 years old. Consumption data were obtained and evaluated with Stata® Statistical Software 16. Different consumption scenarios were considered: the mean Brazilian consumption and the theoretical consumption of different amounts of weekly portions. Body weight was derived from the IBGE survey for Brazilians older than 10 years old (63.6 kg) (IBGE, 2010).

The risk of exposure of the Brazilian population to mercury by the consumption of the analyzed seafood was characterized comparing the estimated methylmercury intakes to the provisional tolerable weekly intake (PTWI) of methylmercury of 1.6 μg/kg body weight per week (JECFA, 2007).

2.4 Statistical analysis

The data was submitted to descriptive statistical analysis, to the Lilliefors test for probability distribution (normality test) and, subsequently, to analysis of variance (ANOVA) and the means were compared by the Tukey test, at 95% probability (Sampaio, 2015), using MINITAB® 18.

3. Results and discussion

3.1. Total mercury levels in shrimps and mussels

The levels of total mercury found in the samples are shown in

Table 1
Total mercury levels in commercial shrimp and mussels.

Seafood	Scientific name	Total mercury levels (mg/kg wb)			
		Mean ± SD	Median	Minimum	Maximum
Whiteleg shrimp	*Litopenaeus vannamei*	0.019 ± 0.014^{b}	0.015	<LOQ	0.071
Atlantic seabob	*Xiphopenaeus kroyeri*	0.011 ± 0.004^{c}	0.010	<LOQ	0.023
Mussel	*Perna perna*	0.031 ± 0.010^{a}	0.028	0.020	0.076

SD – standard deviation (n = 42). wb – wet (weight) basis. LOQ – limit of quantification (0.010 mg/kg).
Mean values in the same column with different letters are significantly different (Tukey test, p ≤ 0.05).

Table 1. All samples presented total mercury below the limit established by the Brazilian and international legislations (0.5 mg/kg for crustaceans, cephalopod mollusks and bivalves) (Brasil, 2013; EC, 2006). Higher total mercury was found in mussels, with values ranging from 0.020 mg/kg to 0.076 mg/kg. These levels are similar to previous studies on *Perna perna* in Brazil (Catharino, Vasconcellos, Sousa, Moreira, & Pereira, 2008; Kehrig et al., 2013; Ansari et al., 2016) and throughout the world (Belabed et al., 2013; Diop & Amara, 2016). Mussels are sessile animals that feed by water filtration, absorbing from it, the nutrients they need to survive but also contaminants (Belabed et al., 2013). Some of these studies were undertaken for environment biomonitoring purpose.

Total mercury levels in the two species of shrimps studied varied from 0.01 mg/kg to 0.07 mg/kg, similar to levels reported in Brazil (Batista, Rodrigues, Souza, Souza, & Barbosa, 2011; Kehrig et al., 2013) and worldwide (Clémens, Monperrus, Donard, Amouroux, & Guérin, 2011; Spada, Annicchiarico, Cardellicchio, Giandomenico, & Di Leo, 2012; Yu et al., 2020). Higher mean total mercury were observed for whiteleg compared to Atlantic seabob shrimp. Factors that can affect mercury are trophic level, age, as well as water and feed contamination (Koenig, Solé, Fernández-Gómez, & Díez, 2013; Rodrigues, Ferrari, Santos, & Conte, 2019). Whiteleg shrimp had higher body weight (15–20 g) compared to Atlantic seabob shrimp (5 g). It is likely that higher mercury levels are found in heavier and bigger shrimps, as reported in the literature (Koenig et al., 2013; Di Lena, Casini, Caproni, & Orban, 2018).

Total mercury levels in the fillet (muscle tissue) and in the exoskeleton (epithelial tissue) of whiteleg shrimps were compared, as both are commonly consumed. Fillet (0.015 ± 0.003 mg/kg) had significant higher total mercury than the exoskeleton (0.009 ± 0.003 mg/kg). Similar results were observed for green tiger shrimp (*Penaeus semisulcatus*) (Kaya & Turkoglu, 2017). This difference can be explained by the higher mercury affinity for the sulfhydryl protein groups – methionine and cysteine, which are abundant in muscle (Houserova, Kuban, Spurny, & Habarta, 2006).

3.2 Influence of heat treatment on total mercury levels in shrimps and mussels

The moisture content and total mercury levels in raw and cooked whiteleg shrimp and mussel samples are indicated in Table 2. There was no significant difference in moisture content between cooked and raw samples for shrimp and mussels; therefore, results were expressed on a wet basis (wb).

Cooking under boiling did not affect significantly total mercury in shrimp and mussels. Previous studies with different types of fish, including tuna, mackerel, shark and edible crab, did not find changes on mercury levels after cooking (Maulvault et al., 2013; Ouédraogo & Amyot, 2011). However, there are contradictory reports, some showing increased (Maulvault et al., 2013; Costa, Korn, Brito, Ferlin, & Fostier, 2016), but others showing decreased mercury in seafood during cooking (Mieiro et al., 2016; Schmidt et al., 2018). Maulvault et al. (2013) observed that cooking affected mercury levels in different ways depending on the species; and reported that the effect of cooking treatments in seafood constituents should be considered in the assessment of risks and benefits.

Table 2
Moisture content and total mercury levels in raw and cooked whiteleg shrimp (*Litopenaeus vannamei*) and South American rock mussel (*Perna perna*).

Cooking time (minute)	Moisture ± SD (g/100 g)	Total mercury ± SD (mg/kg wb)
Whiteleg shrimp		
0	74.5 ± 1.8	0.013 ± 0.007
2	74.2 ± 0.9	0.015 ± 0.008
4	74.2 ± 0.9	0.016 ± 0.010
South American rock mussel		
0	75.8 ± 0.6	0.033 ± 0.003
3	75.0 ± 2.2	0.036 ± 0.006
6	72.5 ± 0.6	0.040 ± 0.012

SD – standard deviation (n = 9). wb – wet (weight) basis.
No significantly difference was observed within a column (Tukey test, $p \leq 0.05$).

Based on this study, mercury in seafood is not significantly eliminated during cooking. This is probably due to the fact that mercury remains bound to proteins (Costa et al., 2016). Even though there were no changes on total mercury levels during cooking, studies are needed to ascertain if heat treatment could affect mercury bioavailability.

3.3. Estimation of Brazilian exposure to methylmercury by shrimp and mussel consumption and potential protective effect from selenium

To estimate exposure to methylmercury from seafood consumption, the weekly exposure was undertaken as recommended by JECFA (2007). There is limited information regarding mercury speciation in shellfish and there was no consensus about the proportion of methylmercury from total mercury, with values varying from 90 to 100% in shrimps and 25–90% in mussels (Batista et al., 2011; Clémens et al., 2011; Spada et al., 2012). Therefore, in this study, a conservative approach was used, assuming that 100% mercury was in the methylmercury form in shrimps and 90% in mussels. Taking these percentages into account, the estimated chronic exposure to methylmercury from the consumption of shrimp and mussels are shown in Table 3.

The mean consumption of shrimp and bivalve mollusks by the Brazilian population was low (0.9 g and 0.1 g per capita per week, respectively) due to the low consumption prevalence: 0.8% and 0.1% of the population reported consumption of shrimp and bivalve mollusks with mean portions of 93 g and 68 g, respectively. For food with low consumption prevalence, it is also necessary to evaluate the risk of the population with a higher consumption profile. Therefore, the methylmercury intake by the consumption of one and three portions per week was considered for the Brazilian population.

The estimated exposure to methylmercury in the different consumption scenarios varied from 0.0003 to 0.09 μg/kg bw per week for whiteleg shrimp, 0.0002–0.04 μg/kg for Atlantic seabob shrimp and 0.00004–0.09 μg/kg for mussels. Even at high consumption (three portions per week), the exposure to methylmercury was low, i.e. the values were only 6% of the PTWI (1.6 μg/kg bw week) established by JECFA (2007).

Mercury in different mussels' species varied widely among studies (Belivermiş et al., 2019; Spada et al., 2012; Yu et al., 2020) and,

Table 3
Estimation of chronic exposure to methyl mercury from the consumption of shrimp and mussel in different scenarios – one and three portions per week.

Consumption scenarios/Seafood	Consumption/capita (g/week)^1	MeHg (mg/kg)^2	Exposure (μg/kg bw)	% PTWI
Mean consumption of Brazilian population				
Whiteleg shrimp	0.9	0.019	0.0003	0.02
Atlantic seabob	0.9	0.011	0.0002	0.01
Mussel	0.1	0.028	0.00004	0.003
One portion per week				
Whiteleg shrimp	93	0.019	0.03	1.7
Atlantic seabob	93	0.011	0.02	1.0
Mussel	68	0.028	0.03	1.9
Three portions per week				
Whiteleg shrimp	279	0.019	0.09	5.2
Atlantic seabob	279	0.011	0.04	3.0
Mussel	204	0.028	0.09	5.6

bw: body weight. MeHg: methylmercury. PTWI: provisional tolerable weekly intake.

[1] Mean portion – shrimp = 93 g and mussel = 68 g (IBGE, 2011).

[2] Estimated value of MeHg: 100% of total mercury for shrimps and 90% for mussels.

sometimes, mussels are reported as an important source of dietary mercury. This was not observed in the commercial mussels *Perna perna*. However, shrimps, in general, are considered a safe seafood option with respect to mercury (Batista et al., 2011; Clémens et al., 2011; Spada et al., 2012; Yu et al., 2020).

The risk associated with mercury exposure by shrimp and mussel consumption was low. However, it can be even lower if the protective effect exerted by selenium is considered (Ralston et al., 2019). In fact, mussel (*Perna perna*) and Atlantic seabob shrimp (*Xiphopenaeus kroyeri*), as well as whiteleg shrimp (*Litopenaeus vannamei*) from the same Brazilian regions were reported to be high sources of selenium (Kehrig et al., 2013; Silva et al., 2016). When using the reported values to calculate the theoretical Se/MeHg molar ratios, values of 20:1 in mussel and values varying from 45:1 to 64:1 in shrimp are obtained. These results reinforce that these shrimps and mussels are safe choices related to mercury and that the health benefits are greater than the risk.

4. Conclusion

The levels of mercury in shrimp and mussels varied among samples, but all of them showed total mercury levels at least 10 times lower than the maximum limits established by the Brazilian and international regulations (0.5 mg/kg). Higher levels of total mercury were observed in mussels compared to shrimps; in whiteleg compared to Atlantic seabob shrimp; and in shrimp muscles compared to the exoskeleton. Culinary heat treatment for up to 4 and 6 min did not reduce total mercury levels in shrimp and mussels, respectively. Estimated methylmercury intakes from the consumption of shrimps and mussels indicate that the studied seafood has low contribution to methylmercury exposure.

CRediT authorship contribution statement

Barbara S. Costa: Conceptualization, Methodology, Investigation, Data curation, Formal analysis, Writing - original draft. **Flávia B. Custódio:** Methodology, Investigation, Writing - review & editing, Supervision. **Valterney L. Deus:** Data curation, Formal analysis. **Daniela C.M. Hoyos:** Methodology, Writing - review & editing. **Maria Beatriz A. Gloria:** Conceptualization, Methodology, Writing - review & editing, Funding acquisition, Supervision, Project administration.

Declaration of competing interest

The authors declare no conflict of interest. The founding agencies had no role in the design of the study; data analyses or interpretation; and in the writing of the manuscript.

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