

# Mercurial Contamination: A Consumer Health Risk Assessment Concerning Seafood From a Eutrophic Estuary in Southeastern Brazil

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Mercury (Hg) contamination has increased in the last decades, resulting in human consumption concerns mainly in developing countries. In this context, this study aimed to carry out a health risk assessment regarding the consumption of swimming crabs, shrimp and squid species caught in different regions of the Guanabara Bay, Rio de Janeiro, Brazil. For this purpose, we used calculations that indicated the Estimated Monthly Intake (EMI), Maximum Monthly Intake Rate (IRmm) and Hazard Quotient (HQ). As the target population, the mean weight corresponding to men and women children aged 12 years, young people aged 24 years, and middle-aged people (adults) aged 54 years were used, taking into account the female and male gender. In the studied seafood, the EMI (0.0001 to 0.0006 mg.kg.month<sup>-1</sup>) was below the monthly intake limit and IRmm (10.3 to 34.8 kg month<sup>-1</sup>) indicates that large quantities of seafood can be consumed by the population studied, unless the safe limit of monthly intake of the contaminant is reached. The hazard quotient (0.4 to 1.4) indicate potential risks health of children eating swimming crabs and squid and young women eating swimming crabs. Our study also highlights the importance of risk assessments, as even when seafood contains Hg concentrations below established limits, consumption variables must be taken into account, so as not to underestimate the potential health risks.

Keywords: crustacean, mollusc, hazard quotient, mercury, Guanabara bay

# INTRODUCTION

Mercury (Hg) contamination in aquatic environments is a serious concern, especially due to the deleterious effects of this metal on the health of seafood consumers, because of its accumulation and toxic effects (Sadhu et al., 2015; Dias et al., 2016; Harayashiki et al., 2018; Rodrigues et al., 2019a; Rodrigues et al., 2019b; Rodrigues et al., 2020). Mercury is a non-essential metal found both naturally in the environment due to volcanic emanations and atmospheric degassing, and anthropogenically, due to pollution especially caused by mining, sewage activity by industrial sectors like chlor-alkalis and metallurgy plants, fossil fuel burning, and domestic sewage (Baptista Neto et al., 2016; Soares-Gomes et al., 2016; Condini et al., 2017; Harayashiki et al., 2018). In Brazil, the Guanabara Bay estuary, located in the state of Rio de Janeiro, is considered an important Hg contamination area, mainly due to the discharges of industrial and domestic sewage. Despite this, it is a prominent artisanal fishing area in the state, due to a very high diversity of species displaying economic value, found even in polluted waters (Baptista Neto et al., 2016; Rodrigues et al., 2019b; Rodrigues et al., 2020).

Mercury is a highly toxic metal, especially in its organic species, methylmercury (MeHg), where it is more bioavailable for absorption and is excreted more slowly (Arcagni et al., 2018). However, regardless of the chemical species, Hg has the ability to bioaccumulate and biomagnify along the entire food chain, being responsible for several deleterious effects on animal and human health (Arcagni et al., 2018; Chen et al., 2018; Mallory et al., 2018; Taylor and Calabrese, 2018).

In humans, Hg effects are noted mainly after chronic exposures, with the dietary route as the main exposure pathway. Among food items, seafood stands out as the main Hg source for humans, due to the wide distribution of this element in the aquatic environment. In this case, both its inorganic form (displaying an absorption rate between 7 to 15%) and organic form (with an absorption rate of approximately 100% of ingested contents) are responsible for intoxication cases (Hong et al., 2012; Rodrigues et al., 2019a). Acute intoxication cases are also linked to elementary mercury exposure, eliminated in mining industries and activities, which is highly volatile (Tchounwou et al., 2012; Jan et al., 2015). In these cases, symptoms are mainly related to somatosensorial and psychiatric disorders that can progress to death (Ekino et al., 2007). Due to the high liposolubility of Hg, especially in the MeHg form, this element easily crosses the bloodbrain and placental barriers, leading to neurological clinical conditions varying in severity, also affecting fetal development. In addition, immunological, cardiovascular and reproductive impairment are also observed (Díez, 2009; Crowe et al., 2017; Gutiérrez-Mosquera et al., 2017; Yin et al., 2017). Chronic intoxication symptoms include cerebellar ataxia, paresthesia of the extremities, somatosensorial disorders, neurological deficits in adults and in children exposed during the prenatal period (Ekino et al., 2007; Bjørklund et al., 2017; Lackner et al., 2018; Rodrigues et al., 2019a). Furthermore, recent studies suggest that Hg exposure may increase an individual's chance of developing Alzheimer's disease (Chakraborty, 2017; Pigatto et al., 2018; Yang et al., 2018; Bjørklund et al., 2019).

As mentioned previously, seafood is the main source of Hg exposure to humans. Many studies have evaluated the role of fish consumption regarding health risks due to Hg exposure (Bi et al., 2018; García-Hernández et al., 2018; Kelly et al., 2018), although others animals such as crustaceans (e.g., swimming crabs, crabs and shrimps) and molluscs (e.g., squid) are very popular in global cuisine and also represent an important Hg exposure risk (Koenig et al., 2013; Liu et al., 2019). This is mainly due to the trophic niche position and habitats of these animals. Crabs and shrimp belong to the lower trophic level, comprising benthic organisms that live in close contact with the sediment, a Hg sink, which can favor high contamination in these animals. Squids are pelagic and animals belonging to higher trophic levels, in some cases, at the same level as fish, as they are predators, and in some cases, due to the effect of biomagnification, can host large amounts of Hg in their bodies (Andrade et al., 2014; Taylor and Calabrese, 2018; Das et al., 2019; Liu et al., 2019). Thus, the assessment of potential health risks due to the consumption of these animals should be further explored.

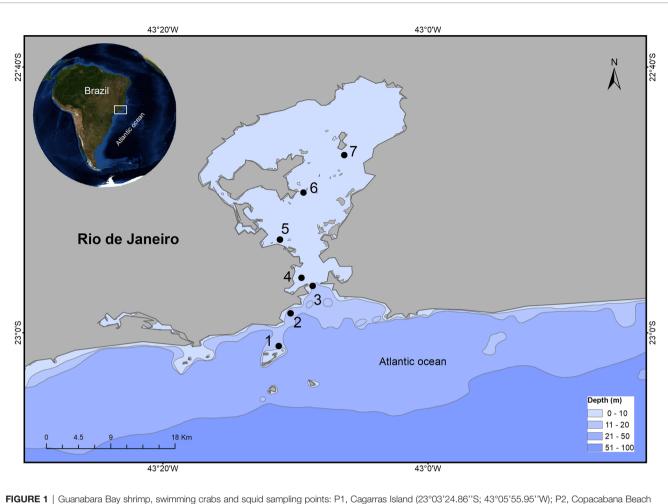
Risks, however, may be underestimated when evaluating only metal concentrations in seafood. Therefore, several factors, such as the consumption frequency of a certain food item, as well as the ingested amounts and exposure period, must also be taken into account. In this regard, the Hazard Quotient (HQ) is noteworthy as an important tool, more accurately representing human health risks, precisely because it considers different factors that interfere in risk occurrence (Copat et al., 2014; Barone et al., 2015; García-Hernández et al., 2018).

Hence, the aim of this study was to carry out a health risk assessment in relation to the consumption of different species of swimming crabs, shrimps and squids captured in different locations in Guanabara Bay, Rio de Janeiro, Brazil. For this purpose, calculations were carried out to assess whether the Hg concentration present in the samples would exceed the tolerable monthly limits, what maximum permitted amount of consumption for the species studied that would not exceed the monthly limit, and the hazard quotient. In this assessment, we simulated the health risk of three populations, children aged 12 years, young people aged 24 years and adults aged between 45 and 54 years, considering men and women separately.

### MATERIAL AND METHODS

#### **Study Area**

All animals were collected by bottom trawls at the Guanabara Bay estuary (22° 24′ – 22° 57′ S, 42° 33′ – 43° 19′ W), located in the state of Rio de Janeiro, southeastern Brazil (**Figure 1**). Seven collection points were chosen, representing the estuarine extension, as follows: Cagarras Island (P1), Copacabana Beach (P2), Urca Beach (P3), Flamengo Beach (P4), Rio de Janeiro seaport (P5), Engenho (P6) and Paquetá Island (P7). A total of 14 bottom trawls (two collections at each point) were carried out at sampling site using a single-port net (25 m net mouth and 6 m high, 25 mm mesh between adjacent nodes). Points were also selected according to the following criteria: presence of port activity and vessel flow (Seaport region), areas of irregular



**FIGURE 1** | Guanabara Bay shrimp, swimming crabs and squid sampling points: P1, Cagarras Island (23°03'24.86''S; 43°05'55.95''W); P2, Copacabana Beach (22°59'24.29''S; 43°09'05.57''W); P3, Urca Beach (22°54'10.12''S; 43°09'05.57''W); P4, Flamengo Beach (22°55'27.10''S; 43°09'02.43''W); P5, Seaport of Rio de Janeiro (22°53'23.73''S; 43°11'03.45''W); P6, Engenho-São Gonçalo (22°50'15.76''S; 43°08'09.97''W); and P7, Paquetá Island (22°47'18.87'S; 43°07'02.23''W).

effluent disposal (present in all sampling points, mainly Engenho and Paquetá), leisure areas and tourist attractions (Cagarras Island, Copacabana beach, from Urca, Flamengo and Paquetá), and local fishing activities (present at all sampling points, especially Copacabana, Urca and the Seaport).

# **Animals Collected**

A total of 125 animals were collected, 48 of which were swimming crabs, 58 shrimps and 19 squids. At each points, we find the following number of animals: 4 at P1 (2 crabs and 2 squids), 17 at P2 (2 crabs, 12 shrimps and 3 squids), 34 at P3 (10 crabs, 21 shrimps and 3 squids), 15 at P4 (6 crabs, 4 shrimps and 5 squids), 28 at P5 (10 crabs and 18 shrimps), 13 at P6 (10 crabs and 3 shrimps) and 14 at P7 (8 crabs and 6 squids) (**Table 1**). After sampling, the animals were taken to the laboratory, identified up to the genus and species, using specialized literature (Crab Database, 2016; FAO, 2018; WRMS, 2019), and dissection of the musculature was performed for Hg quantification. The musculature was chosen as the target tissue, since it is the main tissue destined for food in the three groups of animals.

# **Mercury Quantification**

Before each sample analysis, blank values were verified to be always less than 0.001 Hg (ng). The boats that received the samples were previously dried in a muffle furnace (650°C at 5 minutes) to remove the possible presence of residues from the previous samples. Total Hg in the animals was determined by the

 TABLE 1 | Swimming crab, shrimp, and squid species sampled from

 Guanabara Bay, Rio de Janeiro, Brazil.

Group	Species	n
Swimming crab	Callinectes sapidus	16
	Achelous spinimanus	30
	Achelous spinicarpus	2
Shrimp	Farfantepenaeus brasiliensis	24
	Farfantepenaeus paulensis	12
	Litopenaeus schmitti	22
Squid	Doryteuthis sanpaulensis	16
	Doryteuthis plei	1
	Lolliguncula brevis	2

atomic absorption spectrometry method at Direct Mercury Analyzer (DMA-80), following the manufacturer's recommendations (DMA-80, Milestone, Bergamo, Italy). The equipment was previously calibrated using a standardized Hg solution (Sigma-Aldrich, São Paulo, Brazil). Was prepared a calibration curve from a 1000 mg L<sup>-1</sup> Hg stock, used to build the ten-point calibration curve ranging from 0, 0.5, 1, 2, 3, 5, 10, 20, 50, 100 ng g<sup>-1</sup> (0–100 ng g<sup>-1</sup>; y = 22.085 × x – 0.3217; r 2 = 0.9992). Analytical parameter optimization was performed by limit of quantification (LOQ) and limit of detection (LOD). The LOQ and LOD were determined as recommended by 2011/836/ EU Regulation, and the methodology validation was based on Torres et al. (2012).

Briefly, about 0.2700 g  $\pm$  0.0030 g of each muscle sample were inserted in a quartz boat and dried under an oxygen stream at 160 °C for 1 min, 650 °C for 2 min, and 650 °C for another 1 min, at 3.1 atm. After drying, the Hg vapor was then desorbed using a gold amalgamation trap and released for reading after heating. The detection is performed at a wavelength of 253.7 nm and the results were expressed as mg kg<sup>-1</sup>. Analyses were performed in triplicate.

### **Risk Assessment Calculation**

Calculations were first carried out to determine the Estimated Monthly Intake (EMI) (Equation 1) and maximum monthly Ingestion Rate (IRmm) (Equation 2). The EMI calculation was used to verify whether the Hg concentration present in the average amount of seafood consumed will exceed the permitted monthly consumption limit, established by JECFA (FAO, 2011) and called provisional tolerable mouth intake (PTMI) (0.017 mg kg<sup>-1</sup> month<sup>-1</sup>), which was converted from the weekly PTWI value (0.004 mg kg<sup>-1</sup>). In addition, the IRmm identifies the maximum amount of seafood that can be consumed so as not to exceed the PTMI. Subsequently these data were used to calculate the Hazard Quotient (HQ) (Equation 3). The equations 1 were based on Barone et al. (2015) and equation 3 on Aquino et al. (2017) and followed Environmental Protection Agency guidelines (US-EPA, 1989; US-EPA, 2000). In the HQ equation, values above 1 represent potential damage to consumer health.

$$EMI = \frac{C \times IR}{BW}$$
 Equation (1)

IRmm =  $\frac{PTMI \times BW}{C}$  Equation (2)

$$HQ = \frac{EF \times ED \times IR \times C}{RfD \times BW \times TA}$$
 Equation (3)

EMI: estimated monthly intake; IRmm: maximum monthly Ingestion Rate; IR: ingestion rate (0.093 kg per week and 0.372 per month); C: mercury concentration (mg kg<sup>-1</sup>); BW: body weight; PTMI: is provisorial tolerable month intake (0.017 mg kg<sup>-1</sup> month<sup>-1</sup>); HQ: hazard quotient; EF: exposure frequency (48 days year<sup>-1</sup>); ED: exposure duration (12 or 24 or 54 years); RfD: estimate of a safe daily oral exposure (Hg = 0.0001 mg kg<sup>-1</sup> day<sup>-1</sup>;

US-EPA, 2000); TA: average exposure time for non-carcinogens ("EF" X "ED").

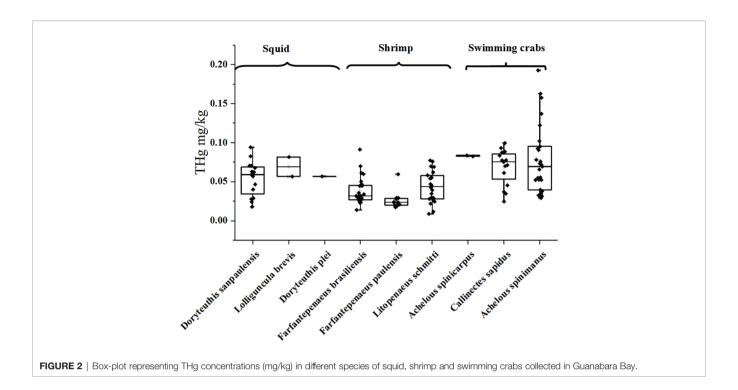
In order to obtain a risk assessment for different age groups and taking into account the differences between men and women, we performed calculations based on the average weight of three age groups: children aged 12 years (boys 42 kg and girls 46 kg), young aged 24 years (men 72 kg and women 59 kg) and adults aged 54 years (men 78 kg and women 66 kg). This information was taken from the platform of the Brazilian Institute of Geography and Statistics (IBGE), which contains data from the Family Budget Survey - Population estimates of the median height and weight of children, adolescents and adults, by sex and age, in the state of Rio de Janeiro.

#### **Statistical Analyses**

Data normality was checked by the Kolmogorov-Smirnov test. The difference between genera and between species was performed using an ANOVA and Fisher's Least Significant Difference (LSD) *post-hoc* test. The Statistica<sup>®</sup> 12 and OriginPro 2020b Trial software programs were used. A significance level of 0.05 was employed for all assessments.

## **RESULTS AND DISCUSSION**

The Hg concentrations detected in the investigated animals were below both Brazilian limits [0.5 mg kg<sup>-1</sup> Hg for non-predator and 1.0 for predator species (ANVISA, 2013)] and international guidelines (0.5 mg kg<sup>-1</sup> Hg) (ANZFA, 2000; FAO, 2003; DOH, 2004; EU, 2005; CSF, 2018; FSA, 2020). The differences between the concentrations of THg in the species studied are identified in Figure 2, however, we did not identify a statistical difference between species of the same type of animal. Mercury concentrations is only statistically different between the three groups of animals studied. Swimming crabs, showed greater contamination which can be attributed to their benthic habitat, in close contact with contaminated sediment. In addition, the bioturbation movement scarred out by these animals result in the resuspension of contaminated sediment takes place, making previously immobilized Hg bioavailable for absorption by the local biota (Das et al., 2019; Liu et al., 2019). When compared to shrimp, which are categorized as the same trophic level, crabs exhibit higher metal assimilation rates and lower excretion rate, resulting in increased Hg values (Evans et al., 2000). Squid were expected to present higher Hg contents due to their predatory habits. This, however, was not observed herein. This can be attributed to the fact that squid are typically pelagic, remaining, consequently, away from the bottom sediment, where Hg is often absorbed. Additionally, Liu et al. (2019) describe that the biomagnification process is more efficient in benthic chains than pelagic ones, corroborating higher Hg concentrations in crabs compared to squids. In addition, physiological and metabolism differences, as well as related to the varied contamination of the Guanabara Bay collection points may also justify this difference (Das et al., 2019; Liu et al., 2019). Other works presented in a literature review by Rodrigues et al.



(2021a) point to the same result found here, indicating that not only for Hg but for other metals, crustaceans, especially crabs and swimming crabs, stand out as important bioaccumulators of these elements, sometimes overcoming the effect of biomagnification (Hosseini et al., 2014; Baki et al., 2018).

In this work we used samples for quantification of Hg, the musculature of the animals. This is because this is the main tissue intended for consumption by aquatic organisms in general (Bisi et al., 2012; Ruus et al., 2017; Arcagni et al., 2018). However, this tissue is not the one with the greatest tropism for the accumulation of metals such as Hg. Rodrigues et al. (2021a) present in their review work a table that points out that, in crustaceans, the main bioaccumulating organ is the hepatopancreas, and secondly the gills, followed by the musculature. In other aquatic organisms, the liver is the organ that performs the function of the hepatopancreas of crustaceans. For this reason, it is also the main accumulator of xenobiotics in fish and cephalopods, such as squid (Azevedo et al., 2016; Mallory et al., 2018; Murillo-Cisneros et al., 2018). This organ stands out for its role in the detoxification process of contaminants, especially for the presence and activity of the metallothionein protein responsible for this mechanism (Azevedo et al., 2012; Hosseni et al., 2013). It has a high percentage of amino, nitrogen and sulfur groups used to sequester metals (Hosseni et al., 2013). On the other hand, despite not being the main site of Hg accumulation, the musculature also plays an important role as a long-term reservoir (Azevedo et al., 2012). This is because when compared to the liver/hepatopancreas, the musculature has a low potential for metal clearance. Even with low concentrations of metallothionein, Hg reaches the muscle through amino acid

thiol ligands that transport it to that tissue (Onsanit and Wang, 2011; Murillo-Cisneros et al., 2018). Thus, based on the importance of muscle consumption, its evaluation is essential, even though it is not the main reservoir of Hg.

Still, on the aspects that can influence the concentration of the contaminant, in addition to the biotic part, there are also abiotic factors, such as water temperature, salinity, oxygenation and pH. These factors were not evaluated in this study; however, based on other previously published studies, it is possible to indicate that such factors are responsible for influencing the bioavailability, speciation, and toxicity of Hg in the Guanabara Bay region (Rodrigues et al., 2019b; Rodrigues et al., 2020; Rodrigues et al., 2021b). The salinity of BG varies between 31 and 35  $g.L^{-1}$ , being closer to the salinity of the marine environment, while the pH is around 8, indicating an alkaline environment (Rodrigues et al., 2021b). Environments with such a salinity rate have a lower concentration of Hg when compared to freshwater habitats. This is because the sulfide is present in saline water complexes with Hg, making it less available for absorption by the biota. As for pH, the bioavailability of Hg is increased in environments where it tends to be acidity (Wang et al., 2010; Dong et al., 2016; Reinhart et al., 2018; Costa et al., 2019). As for BG temperature, varies between 17 and 25°C, and oxygenation between 3 and 6 mg.L<sup>-1</sup> (Rodrigues et al., 2021b). Temperature influences animal metabolism, with higher temperatures responsible for promoting greater excretion of metals such as Hg (Ando et al., 2010). Regarding oxygenation, lower concentrations of oxygen can favor a greater uptake of the same and consequently a greater gill absorption of the contaminants, in addition to favoring the process of methylation of Hg, due to the better activity in this environment of the sulfate-reducing bacteria responsible for the

process (Sadhu et al., 2015). However, most of the characteristics of BG end up disfavoring the acquisition of Hg by the biota due to its low availability for absorption. In addition, a large amount of local organic matter complexes with Hg also reduces its bioavailability (Kehrig et al., 2007). Thus, regardless of the risk factor for consumption, the low concentrations of Hg in BG can be justified by unfavorable abiotic conditions, such as an intensely polluted environment.

The maximum amounts of each seafood species that can be consumed in order not to exceed the monthly intake limit (IRmm) and the EMI values, indicating the concentration of THg consumed monthly through the ingestion of the studied seafood, are shown in Table 2. The EMI results indicate that for all species the THg concentration consumed (93 g per week, 372 g per month) from seafood is very low and below the PTMI limit (0.017 mg.kg.month<sup>-1</sup>). Besides, the results on the IRmm calculation, shown in Table 2, indicate that the estimated amount of consumption allowed to not exceed the provisional tolerable monthly intake for Hg is very high, reaching 34.8 kg for adults and 10.3 for children, demonstrating consumption security, since the quantity that is possibly consumed in the region is very low. General data obtained through statistics available at the Brazilian Institute of Geography and Statistics (IBGE, 2020), indicate that the average per capita consumption of seafood, except fish, in Brazil is 0.2 g day<sup>-1</sup> representing 0.3% of total consumption of these food items. Due to the lack of information in the literature regarding the consumption amount and frequency of the groups of animals investigated herein for the state of Rio de Janeiro, the EMI and HQ calculations were

performed using an amount of 93 g month<sup>-1</sup> of seafood. This value was based on the work by Costa et al. (2021), who indicated the weekly consumption of 93g shrimp by the Brazilian population, a value that we extrapolated to the other animals studied.

Our expectation was that the results obtained in these studies were in line with other results in the literature that demonstrate that, although the concentrations of Hg in these seafood species are below the limit, when considering the amount consumed and the frequency, the calculation would indicate the existence of health risk (HQ>1). In this sense, according to Table 3 we obtained this result when we evaluated the child population of both sexes and women consumers of swimming crabs and squids and young people (24 years old), consumers of swimming crabs. For example, Okati and Esmaili-sari (2017) in their study of fish and shrimp collected in three cities (Bandar Abbas, Bushehr and Mahshahr) located on the coast of the Persian Gulf reported an amount of monthly shrimp consumption (Paeneus semisulcatus and Metapenaeus affinis) (IRmm g month<sup>-1</sup>) of 450 g for children and 990 g for adults, which would result in HQ values of 1.32 and 0.82, respectively. Copat et al. (2014) also identified that, although Hg concentrations in some seafood items from the Gulf of Catania (Ionian Sea) [species: Donax trunculu (crustaceans), Arnoglossus laterna, Mullus barbatu, Engraulis encrasicolus, Trachurus trachurus, Scomber scombrus (all fish)] were within the limit, the intake calculations applying the size of the meals exceeded the doses recommended by JECFA and the HQ values for Hg were greater than 1 in some cases.

Species		EMI						IRmm					THg (mg kg⁻¹ w.w)	n
	Children		Young		Adult		Children		Young		Adult			
	м	w	м	w	М	w	м	w	м	w	м	w		
Shrimp	0.0003	0.0003	0.0001	0.0002	0.0001	0.0002	18.7	20.5	32.2	26.3	34.8	29.5	$0.038 \pm 0.003^{a}$	58
Squid	0.0004	0.0004	0.0002	0.0003	0.0002	0.0003	12.7	13.9	21.8	17.9	23.6	20	$0.056 \pm 0.003^{b}$	19
S. crab	0.0006	0.0005	0.0003	0.0004	0.0003	0.0003	10.3	11.3	17.7	14.5	19.2	16.2	$0.069 \pm 0.003^{\circ}$	43

**TABLE 2** | Maximum consumption rates for squid, shrimp, and swimming crabs sampled from Guanabara Bay, Rio de Janeiro, southeastern Brazil, per month (IRmm in kg month<sup>-1</sup>) and EMI (mg kg<sup>-1</sup> month<sup>-1</sup>).

S. crab, swimming crab; THg, total mercury (kg); EMI, is expressed as mg.kg<sup>-1</sup>.month<sup>-1</sup>; The IRmm is expressed as kg. month<sup>-1</sup>; Concentration: total mercury concentrations (mg kg<sup>-1</sup> w.w) and standard deviations; n, number of samples. Different letters indicate statistical difference between animals (a,b,c)

Body weight- Children (12 years): 42 kg (men) and 46 kg (woman); Young (24 years): 72 kg (men) and 59 kg (woman); Adult (54 years): 78 kg (men) and 66 kg (woman); W, woman; M, men.

TABLE 3 | Results of the Hazard Quotient evaluation in three age groups of the consumer population of squid, shrimp, and swimming crabs sampled from Guanabara Bay, Rio de Janeiro, southeastern Brazil.

Species	HQ								
		Children	Young			Adult			
	м	w		Μ	W	М		w	
Shrimp	0.8	0.7	0.4		0.5		0.4	0.5	
Shrimp Squid	1.1	1	0.7		0.8		0.6	0.7	
Swimming crab	1.4	1.3	0.8		1		0.7	0.9	

HQ, Hazard Quotient; Children (12 years); Young (24 years); Adult (54 years); W, woman; M, men.

When comparing groups comprising different animals, Zhang et al. (2018) identified that fish from major markets in Beijing exhibited higher HQ values than other types of seafood, such as squid and shrimp, which presented HQ values below 1.0. Okati and Esmaili-sari (2017), when comparing fish and shrimp species from different locations within the study region of three cities (Bandar Abbas, Bushehr and Mahshahr) located on the Persian Gulf coast, reported only one HQ value greater than 1.0, regarding shrimp consumption in a population of children and women from fishing communities. Barone et al. (2015) in their study with different species of cephalopods, fish, and crustaceans, identified that none of the groups had values greater than 1.0 in the HQ calculation, and shrimp and squid species from Italy were those that obtained the lowest values. Mishra et al. (2007) also identified HQ<1 values for crabs, praws and bivalves species. These results are in part in agreement with our study, since we did not find values greater than 1 associated with the risk of shrimp consumption and we only identified risk for children consuming squid. In our study, the main risk was associated with the consumption of swimming crabs, since these animals had the highest concentration of Hg when compared to other groups of animals.

Another fundamental aspect to be debated is the difference between the sex and age of the population studied. It is clear that the child population is more likely to develop health problems associated with the ingestion of this contaminant via seafood consumption, since they have a lower body mass, in addition to being with the organism in the development phase (Zamora-Arellano et al., 2017; García-Hernández et al., 2018). These weaknesses may favor the greatest risk to this population. Thus, Zhang et al. (2018) also identified a higher health risk in the 2 to 7-year-old age group with regard to the consumption of certain species of fish, shrimp and squid found in the Beijing market contaminated with Hg. Therefore, the body weight item is an important influencing factor in the risk assessment, since in the evaluated population, the highest HQ values found are in groups with lower body mass, such as children and young and adult women.

Women, mainly in the 20-29 age group, are considered to be of reproductive age. These women, whether pregnant or looking to become pregnant, are sometimes recommended not to consume fish, precisely because it is an important source of metals with genotoxic, carcinogenic and mutagen potential, such as Hg, which has the ability to cross the blood-brain and placental barrier (Ekino et al., 2007; Bjørklund et al., 2017; Lackner et al., 2018). Thus, García-Hernández et al. (2018) in their study identified that women from a fishing community in Kino Bay, Sonora, Mexico were at high risk in relation to MeHg, especially in the age group between 40-49 years, while women between 20-29 years were at lower risk. The study then relates this result to a lower frequency of consumption of seafood in the age group that comprises women of reproductive age, due to this recommendation and a tendency to reduce consumption due to reproduction. Okati and Esmaili-sari (2017) also identified a higher risk for the population of children and women of reproductive age. In this case, such a result was seen in

individuals belonging to fishing communities, where the consumption of this type of food is higher than when compared to the non-fishing population.

Another issue that has not been evaluated and of worth highlighting is the issue of heat treatment to which the seafood is subjected before being consumed. In general, treatments that subject food to heating can lead to direct losses of Hg through volatilization or indirect losses, changing the speciation of this metal to a more volatile form (for example, CH<sub>3</sub>Hg-CH<sub>3</sub>) (Schmitd et al., 2018; Liao et al., 2019). Heat can also lead to changes in solubility and induce metal complexation with other biological components, such as proteins, amino acids or insoluble compounds, thus precipitating during cooking (Maulvault et al., 2011; Schmitd et al., 2018). In addition, several studies indicate that heat treatment can lead to reduced digestibility through changes in conformation or structural losses of the protein associated with Hg. Consequently, these changes can reduce the susceptibility to the action of the protease, decreasing the bioaccessibility of the metal during the gastrointestinal digestion process (Maulvault et al., 2011; Cano-Sancho et al., 2015; Liao et al., 2019). Thus, considering the heat treatment to reduce the Hg concentration in the food and to reduce the bioaccessibility of the metal at the time of digestion, it is possible to indicate that the Hg concentrations that are already low in the seafood studied here, will be lower when subjected to heat treatment, such as cooking, so the health risk may be even lower than the values provided here.

Surprisingly, the Hg risk from consuming seafood was low. It is noteworthy, considering that Guanabara Bay and nearby beaches have water unsuitable for bathing, and several locations in the Bay have intense tailings odors all year round. In this way, contamination by domestic sewage might be being released into this bay in greater quantities, and consequently causing much impacts, when compared to industrial waste.

# CONCLUSIONS

Mercury contamination in crabs, squid, and shrimp from Guanabara Bay is below the limit proposed by Brazilian and international legislation. However, when we consider the average weight of the population, frequency, and quantity consumed, we reveal the risk to the people of infantile and young women consumers of the species with the highest concentration of Hg. Our study reinforces the importance of studying these indicators, not underestimating the risks of Hg consumption when only contamination in seafood is determined. The present study is notable for addressing commercially important but understudied seafood species concerning Hg risk assessments. We also reinforce that, as it is an element that bioaccumulates in the body throughout life, it is not certain that there is no harm to the health of the adult population related to mercury contamination and that the existence of public policies to control contamination by Hg and other pollutants is fundamental in the ecosystem of Guanabara Bay.

# DATA AVAILABILITY STATEMENT

The raw data supporting the conclusions of this article will be made available by the authors, without undue reservation.

### **AUTHOR CONTRIBUTIONS**

PR: Investigation, Formal analysis, Data curation, Writing – original draft, Writing – review and editing. RF: Investigation, Formal analysis, Data curation, Writing – original draft, Writing – review and editing. DR: Formal analysis, Data curation, Writing – review and editing. RH-D: Investigation, Formal analysis, Supervision, Conceptualization, Resources, Data curation, Writing – original draft, Writing – review and editing. LS: Supervision, Conceptualization, Resources. CC-J: Supervision,

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