

Risk map of human intake of methylmercury through fish consumption in Latin America and the Caribbeans

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Abstract

Methylmercury is a high bioaccumulated pollutant persistent in the aquatic environment, that biomagnifies in the food web reaching concerning levels in predator fish. Mining is one of the most important economic activities of Latin America and the Caribbean, and a relevant global anthropogenic mercury emission. Studies have correlated high fish consumption with higher levels of MeHg in humans along with neurotoxic effects. Latin America occupies one of the top 3 regions with the highest fishery exploitation and aquaculture production, and simultaneously, it has been reported in several Latin American and the Caribbean country's high levels of mercury in marine and freshwater fishes, and in human hair associated with fish consumption. Therefore, this review seeks to assess the risk of mercury exposure and consequently health hazard due to fish consumption in Latin America and the Caribbean. The data of mercury levels in fish and fish consumption rates was searched from all countries in Latin America and the Caribbean. A large data set was created evidencing a concerning presence of methylmercury in fish that are widely consumed. The 6.1% of the total fish species studied were found to have concentrations of $\text{MeHg} \geq 1.5 \mu\text{g g}^{-1} \text{ dw}$, independently of the water habitat these were mainly carnivore species, which is recommended to not consume. Furthermore, high risk values ($\text{HQ} \geq 1$) were estimated in Peru and Venezuela, and even higher-risk values ($\text{HQ} \geq 10$) were estimated in some fish species inhabiting watersheds in Trinidad. The recommendation is to lower the consumption of this kind of fish species or to avoid mercury pollution.

Introduction

The metallic element mercury (Hg) in its different chemical forms is widely distributed in the environment. It is found in all kinds of ecosystems due to its many natural sources such as volcanic emission, soil and rocks erosion, wildfire, but also due to anthropogenic activities such as fossil fuel burning and mining extraction (Pirrone et al., 2010). Aquatic ecosystems are one of the most relevant for the Hg cycling from the atmosphere due to its methylation carried out by microorganisms in aquatic sediments, giving rise to methylmercury (MeHg), an organic form of Hg with higher toxicity

MeHg is persistent in the environment, reaching between 10–30% of the total Hg (THg) in the aquatic environment (Mason et al. 2000) and its bioaccumulation occurs through the food chain. In fact, more than 85–90% of the Hg in fish is MeHg and its concentration has shown to be correlated with the trophic position, age, and size (Grieb et al. 1990; Southworth et al. 1995). Therefore, humans are susceptible to MeHg's harmful effects by the intake of MeHg through fish (Rice et al. 2014; Lee et al. 2020; Guzzi et al. 2021), particularly those predator species with the highest trophic position in the food chain (Langeland et al. 2017).

Top predator fish are found to have very high THg levels (EPA's contamination level $0.46 \mu\text{g g}^{-1}$ wet weight or $1.5 \mu\text{g g}^{-1}$ dry weight) (Lasut et al. 2010; Esdaile and Chalker 2018). Latin American and the Caribbean produced 18% of global anthropogenic Hg emissions, of which small scale gold mining contributed 70% (340 t out of 2,220 t). In 2015, South America alone accounted for 53% of the estimated

1,220 t of Hg released globally from small scale gold mining to aquatic and terrestrial environments (UN Environment 2019; Canham et al. 2021).

In addition, Latin American and the Caribbean occupies one of the top 3 regions with the highest fishery exploitation and aquaculture production (FAO 2020). Indeed, it has been reported significant mercury concentrations in marine and freshwater fish, and in human hair associated with fish consumption in several Latin American and the Caribbean countries (Langeland et al. 2017; Bravo et al. 2019; Valdelamar-Villegas and Olivero-Verbel 2020).

Neurotoxic and motor skill effects in humans have been related to high MeHg concentration accumulation ($\geq 1 \mu\text{g g}^{-1}$ dry weight; dw) (ATSDR 1999; Guzzi et al. 2021). One of the most well-known cases is the Minamata disaster in the 50's at Minamata Bay in Japan, where there was a high scale poisoning with fish and sea food contaminated with MeHg. This compound was generated as a by-product in reaction chambers for manufacturing acetaldehyde and discharged directly to the bay accumulating in fish and seafood that later was consumed by the local inhabitants. The intake of contaminated marine products produced a syndrome called Minamata Disease, consisting in sensory disturbance in the extremities, ataxia, disequilibrium, bilateral concentric constriction of the visual fields, impairment of gait and speech, muscle weakness, tremor, abnormal eye movement, and hearing impairment. Mental disorder and disturbances of taste and smell are also present occasionally (Eto 2000; Hachiya 2006). Besides this high exposure levels to MeHg, chronic exposure to lower levels can cause hepatic diseases and reproductive toxicity (Tan et al. 2009; Rice et al. 2014). In 2013 the Minamata Convention was created and adopted by 137 countries until today, of which 20 are countries from Latin America and the Caribbean (Hachiya, 2006). Therefore, it is of major importance to implement a risk assessment method that can be applied in decision making and in the generation of environmental regulations in vulnerable countries such as those of Latin-American and the Caribbean. This literature review and meta-analysis seeks to assess the risk of Hg exposure and consequently health hazard due to fish consumption in Latin America and the Caribbean by collecting available data from these countries regarding Hg levels in fish and fish consumption rates in each country.

Methods

Literature review

A literature search on peer-reviewed journal articles and published reports documenting Hg concentrations in fish in Latin America and the Caribbean countries was conducted. Studies published from 2010 until 2022 were selected, with some exceptions of countries where the data were only before 2010. The search was performed in Web of Science and Google Scholar using all combinations of the following keywords: "mercury," "fish" AND "Latin America"; "mercury", "human" AND "Latin America"; "mercury", "fish" AND "country" (every Latin American and the Caribbean country separately); and "mercury", "human" AND "country" (every Latin American and the Caribbean country separately)".

Data was classified by country, site (GPS), habitat (marine or freshwater environment) and fish diet (Supplementary Table S1) by fish species. In addition, the Hg species measured (THg and/or MeHg) and the analytical methods used were also considered in the classification.

Mercury risk assessment

The data was filtered by (1) fish species that are consumed by humans, and (2) Hg concentration quantified in muscle that corresponds to the most consumed fish tissue.

Additional data was necessary to determine the level of health risk in the population of each country. Average body weight for adult females (BWF) and males (BWM) from each country was obtained from Eglitis dataset (Eglitis 2021) and average fish consumption per country and per day (FCR) was obtained from the FAO report (FAO 2020) (Supplementary Table S1).

The Target hazard quotient (THQ) is a non-carcinogenic index that assesses the health risk from food consumption by residents and is calculated by equation (Formula 1) (Felix et al. 2022).

$$THQ = \frac{EF \times ED \times FCR \times C_{Hg}}{RfD \times BW \times AT} \times 10^{-3} \text{ (Formula 1)}$$

Where EF is the exposure frequency (365 days/year). The factor ED is the total duration of fish consumption (average human life 70 years), FCR is the fish consumption rate (g per day), C_{Hg} is the mercury concentration expressed as ($\mu\text{g g}^{-1}$ wet weight), BW is human body weight (Kg), AT is the average time of food consumption (EF x 70 years), and RfD is the oral reference doses as (0.001 mg per Kg per day) (EPA 2011).

Recommendations for fish consumption (avoid, keep and encourage consuming) were based on the work done by Vieira et al. 2021, and the following equation (Formula 2):

$$MPW = \frac{(RfD \times BW) \times 7 \text{ days}}{(Fish \text{ meal size (g)} \times C_{Hg}) / 1000} \text{ (Formula 2)}$$

For the size of the fish meal the 2015–2020 Dietary Guidelines for Americans who recommend at least 226.8 g of fish per week based on a 2000 cal diet was used (Dietary Guidelines Advisory Committee 2015), and a body weight (BW) was based on the adult population average specific by country and human sex (BWF: female and BWM; male).

Data analysis

The data are presented as MeHg concentration mean \pm SD ($\mu\text{g g}^{-1}$ dw). Due to Shapiro-Wilk's test determined a non-normal distribution of the data, non-parametric statistical tests were used to estimate statistical differences ($p = 0.05$). Kruskal-Wallis and Dwass-Steel-Critchlow-Fligner pairwise comparison tests were employed to determine the mean differences in MeHg concentrations among trophic levels, habitat type (marine or freshwater), and countries (Supplementary Table S2).

A subsample corresponding to the highest quartile of distribution based on the mercury content was used to visualize the frequencies of habitat type (marine or freshwater) and diet preferences (carnivores, detritivores, herbivores, or omnivores) within the fish species. With these classifications a Sankey plot was built (Fig. 1) using the *Sankey Network* function from the *networkD3* R-package (Allaire et al. 2017).

To compare the risk (HQ) differences between human sex (female and male population), a Mann-Whitney test was performed (Supplementary Table S3). To observe the geographic distribution of the Hg exposure risk by fish consumption among inhabitants of Latin America and the Caribbean countries, the obtained risk values for women and men were mapped using the geographic coordinates reported in the respective paper of origin. For this, the *rgdal* (Bivand et al. 2021) and *ggplot2* (Wickam 2016) R-packages (R Core Team 2021) were used.

All plots resulting from the information gathered were created with *ggplot2* (Wickam 2016) and R-packages (R version 4.1.2).

Results

Mercury concentration on fish

Studies concerning Hg bioaccumulation in fish consumed by humans were found around 30% of Latin-American and the Caribbean countries including, from north to south: Jamaica, Suriname, Anguilla, Trinidad & Tobago, Cuba, Mexico, Nicaragua, Costa Rica, Venezuela, Colombia, Peru, Brazil, Bolivia, Argentina, and Chile. Brazil and Chile lead in investigation around Hg bioaccumulation in fish. Most of the fish species studied belong to freshwater ecosystems, with no data available for marine fish in Bolivia, Cuba, Peru, and Venezuela. A total of 278 species were analyzed for THg or MeHg in muscle, where 61.2% belong to freshwater ecosystems and 38.8% are from marine ecosystems. Most of the data belong to wild capture fishes.

Freshwater fish MeHg concentrations range goes from 0.02 to 2.1 $\mu\text{g g}^{-1}\text{dw}$; meanwhile marine fish range goes from 0.01 to 10.7 $\mu\text{g g}^{-1}\text{dw}$ (Table 1), the tests showed no significant differences between the bioaccumulated MeHg between habitats (p -value = 0.585; Supplementary Table S2). Although marine fishing is an important economic activity, there is less information about mercury pollution on marine fish.

Higher MeHg concentrations ($\geq 1.5 \mu\text{g g}^{-1}\text{dw}$ of MeHg) were found in mostly carnivore species, except for *M. acanthogaster* ($1.5 \mu\text{g g}^{-1}\text{dw}$) an herbivore specie from Venezuela. The highest MeHg concentrations are found in the marine species *C. pororus* ($11.1 \mu\text{g g}^{-1}\text{dw}$) and *S. lewini* ($6.3 \mu\text{g g}^{-1}\text{dw}$) from Trinidad. Meanwhile the freshwater species with the highest MeHg levels are the predator *O. mykiss* ($2.1 \mu\text{g g}^{-1}\text{dw}$) from Chungara catchment in Chile and *C. monoculus* ($1.8 \mu\text{g g}^{-1}\text{dw}$) from Brazil (Table 1). These concentrations are considered highly hazardous to health by the FAO regulations (FAO and WHO 1995).

The Sankey plot (Fig. 1) shows a subsample of those fish species from the highest 20% MeHg concentration. This includes 57 fish species with a MeHg concentration $\geq 0.5 \mu\text{g g}^{-1}\text{dw}$. The remaining 80% is considered to have concentrations that are not harmful and are safe to be consumed in a moderated diet. Various species are repeated in the list because: a) same species were found in different countries, and b) same species were sampled in different sites within the same country. The MeHg concentration is represented by the width of grey ribbons. Dark or light blue bars indicate the distribution between water habitats (marine or freshwater). Food preferences (carnivorous, omnivorous or herbivore) are indicated by colored bars to the right. The plot showed that carnivorous species present the largest range and highest concentrations of MeHg (0.01 to $10.7 \mu\text{g g}^{-1}\text{dw}$), independently of the water habitat. However, one omnivorous (*C. gariepinus*, $1.1 \mu\text{g g}^{-1}\text{dw MeHg}$) and one herbivore specie (*M. acanthogaster* $1.5 \mu\text{g g}^{-1}\text{dw MeHg}$) also displayed MeHg concentrations that might deserve attention since most of the food intake recommendation are below $1.5 \mu\text{g g}^{-1}\text{dw}$ of Hg.

Health risk of MeHg bioaccumulation in fish from Latin America

The Venn diagram (Fig. 2) summarizes the distribution of MeHg concentration and its interaction with THQ values in the total species assessed in this study. The 93.9% of the total species present MeHg concentrations $< 1.5 \mu\text{g g}^{-1}\text{dw}$, from which 8.6% contributed to THQ values ≥ 1 . The remaining 6.1% of the species with MeHg levels $\geq 1.5 \mu\text{g g}^{-1}\text{dw}$ contributed in its totally to the THQ values ≥ 1 , 1.4% to THQ ≥ 5 and a 0.7% to THQ ≥ 10 . This group of fish species with MeHg concentrations above the FDA/EPA recommended levels ($\geq 1.5 \mu\text{g g}^{-1}\text{dw}$) are found in Trinidad, Colombia, Chile, Brazil and Venezuela, and would not to be recommended for frequent human consumption.

The geolocation of the health risk estimation is presented in Fig. 3. The level of human fish consumption is indicated by color, where high consumption fish are those consumed locally and of highly commercial interest (orange), while the low consumption fish are consumed only locally (green). Additionally, considering that a THQ ≥ 1 is considered hazardous for human health, the frequency plot shows that most studied fish species have no risk for human consumption (85% for female and 86% for male).

In summary, fish species showing MeHg concentrations $\geq 1.5 \mu\text{g g}^{-1}\text{dw}$ and express THQ ≥ 1 ; are those corresponds to fish species from Anguilla, Brazil, Chile, Colombia, Jamaica, Nicaragua, Paraguay, Peru, Venezuela, and Trinidad. Yet, fishes with lower MeHg concentrations ($< 1.5 \mu\text{g g}^{-1}\text{dw}$) also show to have an HQ ≥ 1 in almost every country (Supplementary Table S1). All species studied from Argentina (20), Bolivia (18), Nicaragua (18), Costa Rica (7), Cuba (7), Ecuador (2) and Mexico (24) show an HQ < 1 .

There is a slight trend difference in the risk estimation regarding human female and male population, being the female population the one with the higher THQ values, however no statistical difference between both groups was found (Fig. 3, p-value = 0.268, mean difference 0.07, Supplementary Table S3).

The countries estimated to have the greater health risk were Peru and Venezuela due to more than 50% of the species studied from these countries present a $THQ \geq 1$. Fewer species contributed to very high THQ values ($THQ < 10$) as *C. porosus* (HQF = 32.1; HQM = 29.9) and *S. lewini* (HQF = 18.3; HQM = 17.1).

Discussion

There is a lack of information regarding bioaccumulation of Hg in wildlife and aquaculture fish. Most countries considered in this research do not have an actualized survey of the state of contamination of their fish species. This is a matter of concern since mining is one of the most important economic activities in the history of Latin America (Alimonda 2015) and one of the most relevant Hg contamination sources (Esdaile and Chalker 2018). In spite that most of the global fishery and aquaculture activities are centered in marine species (FAO 2020) there is lack of information of Hg amounts in these species, i.e., 83% of Chilean fishery exports comes from marine fisheries whose main export species are *Trachurus murphyi*, *Engraulis ringens*, and *Strangomera bentincki* and only one of them is mentioned in the studies (SUBPESCA 2020; OECD 2021)

One of the regions responsible for Hg emissions, east and southeast Asia, also characterize for a high mining and industry economy (UN Environment 2019; Soe et al. 2022) studies from these countries have shown a lower average Hg bioaccumulation in fish that some found in Latin America and the Caribbeans (Hajeb et al. 2009; Park et al. 2011; Jeevanaraj et al. 2019; Shalini et al. 2021; Soe et al. 2022).

The risk index THQ values obtained in this study (HQF = 0.02–32.1; HQM = 0.01–29.9) show to have a wider range and extremely high THQ values compared to other studies such as the one done by Bonsignore et al. (2013) in Augusta Bay in southern Italy ($THQ = 0.31–15.8$); also, Azore archipelago $THQ = 0.1–8.6$ and $THQ = 0.2–8.5$ on the Northwest Portuguese (Felix et al. 2022). The higher MeHg level were dominated by Carnivore species which is consistent with previous studies (Chiang et al. 2021), due to MeHg biomagnification through trophic levels and carnivores are on the top of the trophic web accumulating more MeHg than lower levels. These same species were found to have a significant contribution in the estimation of risk finding a relation between high MeHg concentrations and $THQ \geq 1$ value. Nevertheless, in some cases high MeHg levels were not necessarily the cause for high HQ values. This was observed in Peru where all species were found to have safe MeHg concentrations ($< 1.5 \mu\text{g g}^{-1}\text{dw}$), although the THQ values were ≥ 1 . Particularly in Peru, they have a high fish consumption rate (68.6 g day^{-1}) of this country due to its gastronomic preferences, compared for example to Argentina a country whose fish species have similar MeHg levels to Peru's, but its fish consumption rate is between the lowest in Latin America and the Caribbeans (15.6 g day^{-1}) therefor Argentina THQ values are safe. Meanwhile, in countries like Trinidad and Jamaica most of their fish species have acceptable MeHg levels, still they have HQ values ≥ 1 . This exemplifies that regardless of having safe MeHg concentrations, the fish consumption rates ($> 65 \text{ g day}^{-1}$), and body weight ($> 76 \text{ Kg}$ per females' adult and $> 80 \text{ kg}$ per male adult) are important factors to determine the safety of fish for human consumption. Therefore, it would be advisable to consider demographics and cultural behavior factors in the design of

the regulations and health safety guidelines and not only on the concentrations of Hg found in the fishes' muscle.

For which in Table 2 there is a recommendation chart for those species to avoid consuming, keep consuming with moderation and to encourage their consumption based on their MeHg levels and THQ values obtained during this study. This classification is based on the MPW (meals per week) index, where < 1 MWP is recommended to avoid, 1 > 2 MPW to keep consuming with moderation and < 2 MPW to encourage their consumption, this is specified by country.

Conclusion

As a result of this study, it is possible to conclude from the total species studied in this research 47% are considered safe and non-risky to consume at the same rate that they are being consumed. Likewise, it is recommended to avoid consuming fishes from some genus such as *Thunnus* and *Epinephelus* due to the high MeHg concentrations, and in the case of fishes with concentration below $1.5 \mu\text{g g}^{-1}\text{dw}$ to reduce the consumption of them to minimize the health risk.

As a suggestion, because of the level of mining activity developed in Latin America and the Caribbeans, these countries must improve environmental regulations to help to prevent population health issues. It is not possible to regulate under other countries' standards due to the differences in distribution of Hg emission, population behavior and demographic characteristics.

Declarations

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Ethical Approval 'Not applicable'

Consent to Participate 'Not applicable'

Consent to Publish 'Not applicable'

Authors Contributions

All authors contributed to the study conception and design. Material preparation, data collection and analysis were performed by the authors E.V. and L.Z. The first draft of the manuscript was written by E.V. and all authors commented on previous versions of the manuscript. All authors read and approved the final manuscript.

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Competing Interests

The authors have no competing interests to declare that are relevant to the content of this article.

Availability of data and materials

The data that support the findings of this study are available from the corresponding author, P.B., upon reasonable request.

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Tables

Table 1 and 2 are available in the Supplementary Files section.

Figures

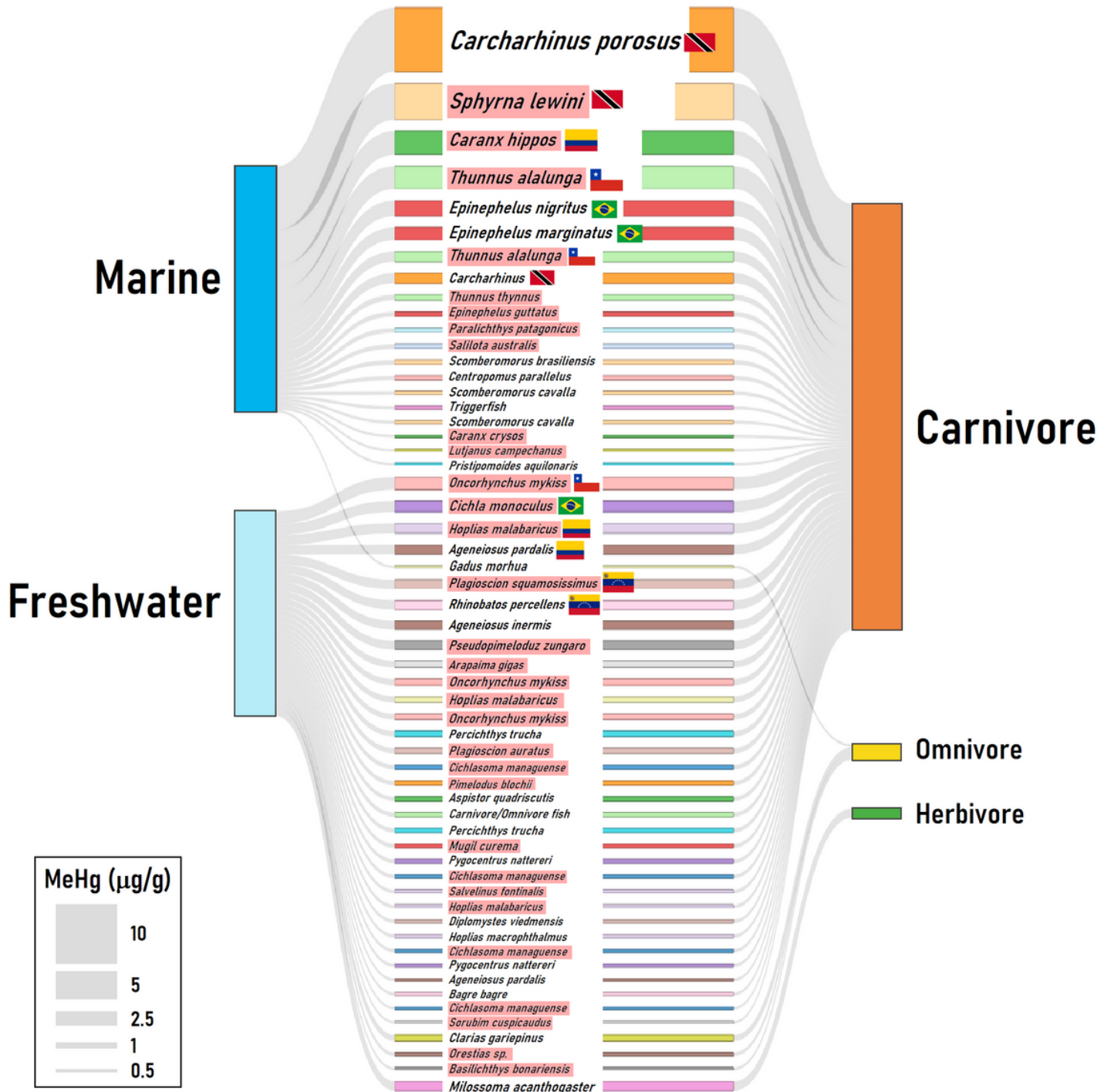


Figure 1

Sankey diagram representing the highest 20% of MeHg levels in fish muscle from Latin-America and the Caribbean. In red are highlighted those species most consumed. Species with MeHg concentrations above the FDA/EPA recommended levels ($\geq 1.5 \mu\text{g g}^{-1}$ dw) are highlighted with the country of origin's flag.

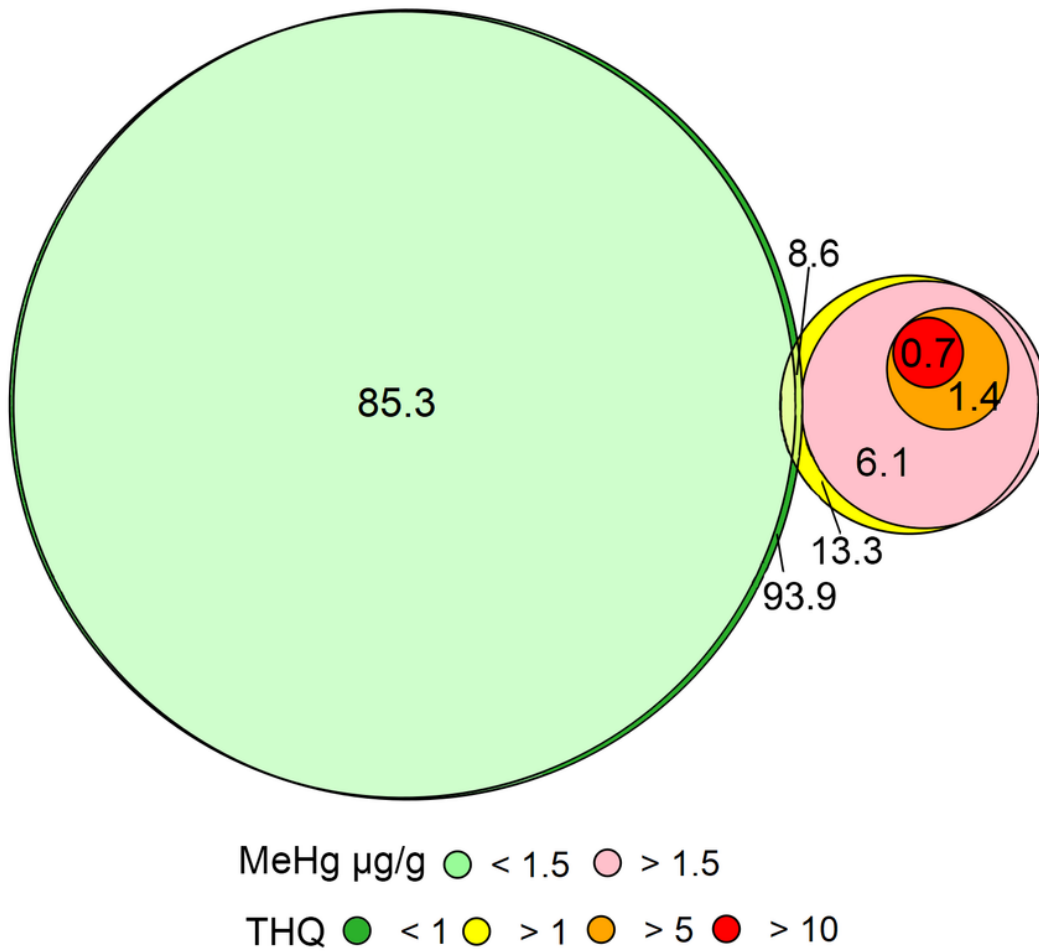


Figure 2

Percentage of MeHg and THQ values, and their association within the species assessed.

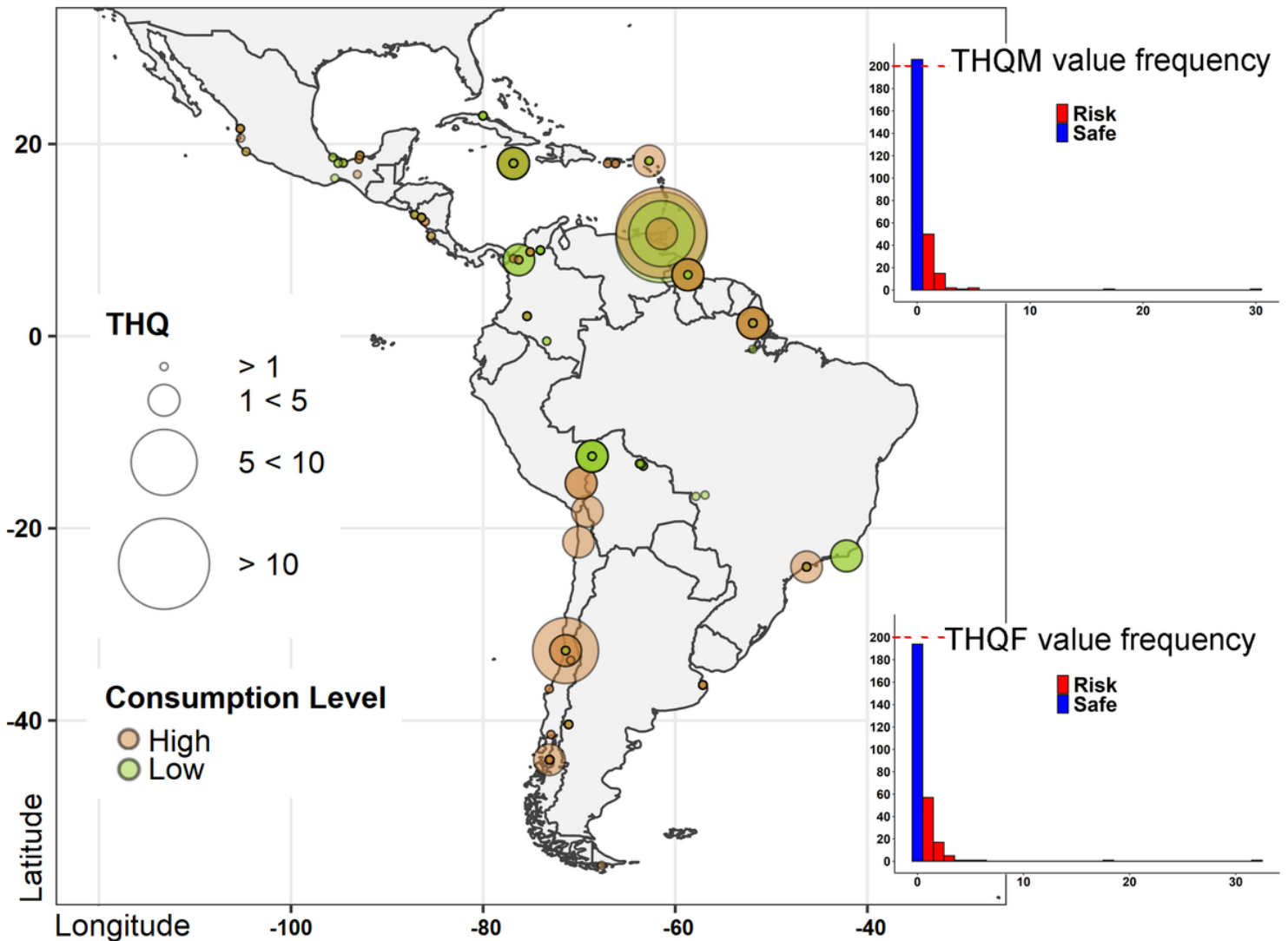


Figure 3

Estimated risk map for human population related to MeHg contaminated fish consumption from Latin American and the Caribbeans. Circles sizes indicate the average THQ value. The color indicates if the specie is highly consumed (orange) or locally consumed (green). The bar graphs illustrate the frequency of THQ for human male population (THQM, up) and female population (THQF, down). Also, THQ > 1 (red bar) and < 1 (blue bar).

Supplementary Files

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