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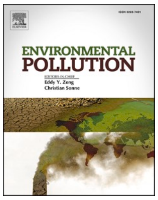

Review

Mercury in oceanic upper trophic level sharks and bony fishes - A systematic review

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ABSTRACT

Anthropogenic activities contribute to nearly half of current Hg emissions to the atmosphere. In the marine habitat, oceanic predator fishes bioaccumulate Hg throughout their lives, making their consumption the main route of Hg exposure in humans. In this context, several publications, between 1973 and 2022, were selected, analyzed, and duly compiled, with the objective to investigate Hg contamination in nine species of bony fish: *Thunnus thynnus* (8 publications), *Thunnus albacares* (19), *Thunnus obesus* (7), *Thunnus atlanticus* (5), *Thunnus alalunga* (4), *Katsuwonus pelamis* (8), *Xiphias gladius* (18), *Coryphaena hippurus* (7) and *Euthynnus alletteratus* (4), as well as two species of cartilaginous fishes *Prionace glauca* (13 publications) and *Isurus oxyrinchus* (8). These studies totaled 5973 individuals. We classified species according to taxonomic groups and region of capture and found a significant difference between sharks and bony fishes, with higher Hg concentrations in sharks. The regions of occurrence were divided into 4 large areas (North Atlantic - NAO, South Atlantic - SAO, Equatorial Atlantic Ocean - EAO, and Mediterranean - MED), but no significant differences were observed when comparing the overall Hg concentrations in fish among regions (including all species). Additionally, a thorough discussion of the risks associated with human consumption of these species was conducted, as nine of the selected species presented individuals with Hg concentration values that exceeded the safety limits (1 ppm) set by health agencies worldwide.

1. Introduction

Concern with metal contamination is not recent as scientific research addressing such issue have been developed since the beginning of the last century. Within this class of contaminants, mercury (Hg) stands out mainly due to its toxicity and long-term occurrence in the environment (Hintelmann, 2010). From a combination of natural (mostly of volcanic and degassing origin) and anthropogenic (mostly from fossil fuel combustion, cement production and artisanal gold mining) sources, Hg is emitted mainly to the atmosphere from here can be deposited on ocean surfaces and incorporated into marine food chains (UNEP, 2018). Anthropogenic sources contribute to nearly half of the total global Hg emissions altering its biogeochemical cycling (Driscoll et al., 2013; Stern et al., 2012; Gworek et al., 2016; UNEP, 2018). In pelagic oceanic regions, atmospheric deposition is the dominant source of Hg, mostly as Hg^{2+}, which can be methylated in the photic zone and forming methylmercury. Exception are the Arctic Ocean where riverine inputs are more important than atmospheric sources (Obrist et al., 2018; Soerensen et al., 2010).

Mercury (Hg) is a toxic element to living organisms in general, including humans, and aquatic environments are the main route of Hg exposure to humans and wildlife. The inorganic Hg is transformed by bacteria-mediated reactions into methylmercury, its most toxic form, that efficiently accumulates in aquatic organisms that can subsequently be consumed by humans (UNEP, 2018). Events of Hg intoxication in humans, commonly associated with exposure to high doses of Hg, are reported worldwide with neurological problems being the most reported symptom. However, long term exposure to relatively low doses may also result in changes of vital functions, including behavior, reproduction and physiology (Clarkson, 2002). The first officially reported mercury poisoning accident was in Minamata, Japan, in May 1956, where an outbreak of Hg poisoning in fish was reported for the first time. Mercury present in the wastewater of a chemical plant (Chisso Co. Ltd.) contaminated the local seafood resulting in severe poisoning of human

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consumers (Harada, 1995). Nearly six decades later, Hg concentration in many edible fish species in the Minamata bay still exceeds the Japanese regulatory limits (Kindaichi and Matsuyama, 2005; Yoshino et al., 2020), highlighting the long-lasting effects of Hg contamination and the challenge to restore areas following Hg contamination incidents.

Studies on dietary Hg assimilation in animals report several dysfunctions in the endocrine, reproductive, and immune systems, even when exposed to trace amounts of Hg (Schneider et al., 2013; Spalding et al., 2000). Characteristics such as size, age, habitat, trophic position, and life span are factors known to influence the magnitude of the accumulation, partitioning and internal distribution of Hg in fish (Schneider et al., 2013). For example, large-bodied fishes of upper trophic levels generally present higher concentrations of Hg compared to lower trophic level fishes (Andersen and Depledge, 1997; Mendez et al., 2001; Storelli et al., 2002). This pattern is commonly observed in tuna, tuna-like and shark species, which are upper-level predators of oceanic food webs, typically presenting more than 95% of their Hg burden as methylmercury (Monteiro and Lopes, 1990; Storelli et al., 2001; UNEP, 2018).

In the Pacific Ocean, diet, feeding behavior, and habitat, as well as variations in environmental Hg levels, determine the Hg body burdens in tuna species (Houssard et al., 2019). However, data from other regions of the world are still scarce. In the Atlantic Ocean, for example, recent studies suggest significant differences in Hg concentrations in different sectors of the Equatorial Atlantic region (Lacerda et al., 2017), but a detailed description of the relationship between Hg accumulation and physiological, ecological, and environmental parameters is still lacking for the Atlantic Ocean.

The world fish production has been increasing decade by decade, reaching 178.5 million tons in 2018, with 156.4 million tons destined for human consumption (FAO, 2020). It appears that the world population is seeking a diet with more fish, as per capita consumption is increasing (20.5 kg year^{-1}) (FAO, 2020). Tuna species such as, Skipjack and Yellowfin (included in this review) are among the top 10 most caught of world's fisheries (FAO, 2020). In this context, sharks, tunas, and associated species are the focus of the present review, because besides being widely distributed in world's oceans, which render comparability, they are also caught for human consumption (Hazin and Travassos, 2007; FAO, 2020). In the present study, we analyze Hg contamination in top pelagic predators (sharks and bony fishes) caught in the Atlantic and Mediterranean aiming to better understand patterns among ocean areas, taxon, and other relevant biological and ecological traits that influence Hg concentrations in these fishes. In addition, we provide a risk assessment of the consumption of these species by the general population.

2. Material and methods

The PRISMA (Preferred Reporting Items for Systematic Reviews and Meta-Analyses) protocol were used to conduct this systematic review (Liberati et al., 2009; Moher et al., 2016). We selected publications reporting total mercury (Hg) concentrations in nine tuna and tuna-like species, including Bluefin tuna (*Thunnus thynnus),* Yellowfin tuna (*Thunnus albacares),* Bigeye tuna *(Thunnus obesus),* Blackfin tuna *(Thunnus atlanticus),* Albacore (*Thunnus alalunga),* Skipjack tuna *(Katsuwonus pelamis),* Swordfish *(Xiphias gladius),* Dolphinfish (*Coryphaena hippurus),* and Little tunny *(Euthynnus alletteratus)*, and two shark species, including Blue shark *(Prionace glauca)* and Mako shark (*Isurus oxyrinchus)* from the Atlantic Ocean and the Mediterranean Sea.

We searched the scientific literature databases, using institutional access provided by the Coordination for the Improvement of Higher Education Personnel (CAPES) of the Brazilian Ministry of Education, including Web of Science, Science Direct, Scielo, and Google Scholar. Searching terms and keywords were used individually and/or in combination and included all species scientific names, in addition to *"tuna", "sharks", "swordfish", "blue shark", "mako shark", "albacore tuna", "skipjack tuna", "bluefin tuna", "bigeye tuna", "yellowfin tuna", "blackfin tuna", "little tunny", dolphinfish, Hg, mercury, "Atlantic Ocean", Atlantic, and/or Mediterranean.* Publications included scientific articles, theses, and dissertations available on digital databases published from 1973 to mid-2022 in the English, Spanish and/or Portuguese language. Undergraduate monographs, abstracts, and articles not available on digital databases were not included.

The information recorded from selected scientific articles included year of publication, species, sample size, legal limits, oceanic location (North Atlantic Ocean - NAO, South Atlantic Ocean - SAO, Equatorial Atlantic Ocean - EAO and Mediterranean - MED), body size, body weight, mean Hg concentrations in muscle tissue, standard deviation, minimum and maximum Hg concentrations, and bibliographic references. Subsequently, species were also grouped into Class (Osteichthyes and Elasmobranchii), Order (Perciformes, Carcharihiniformes, and Lamniformes), Family (Scombridae, Carcharhinidae, Coryphaenidae, Xiphiidae, Lamnidae) and Genus (*Thunnus, Katsuwonus, Prionace, Coryphaena, Xiphias, Isurus*, and *Euthynnus*).

Mercury concentrations were expressed in nanogram per gram (ng g^{-1}) on wet weight basis (w.w.) for muscle tissue only. When necessary, transformations from dry weight to wet weight were performed using the moisture content reported in the respective study. The criteria adopted to assess the exposure risk to humans through the consumption of seafood were the established maximum acceptable limit of 1000 ng g^{-1} for these predator species according to the World Health Organization (FAO/WHO, 2011).

2.1. Statistical analyses

Shapiro Wilk test for normality followed by the non-parametric Kruskal-Wallis were applied to identify potential differences in Hg concentration between factors, such as taxa (species, order, family, genus), oceanic areas, and year of publication. All test analyses and graphing were conducted using Microsoft Excel and R Software (R Core Team, 2017).

3. Results and discussion

3.1. Overall publications and Hg concentrations

Several publications, between 1973 and March 2022, were selected, analyzed, and duly compiled. The analyzed papers totaled 5973 individuals among the selected species, and the following number of publications per species: 8 *T. thynnus,* 19 *T. albacares,* 7 *T. obesus,* 5 *T. atlanticus,* 4 *T. alalunga,* 8 *K. pelamis,* 18 *X. gladius,* 7 *C. hippurus,* 4 *E. alletteratus* and finally 13 *P. glauca* and 8 *I. oxyrinchus* (Fig. 1).

The highest concentrations, on average, were observed in the sharks *I. oxyrinchus* (988 ± 693 ng g^{-1}) and *P. glauca* (970 ± 607 ng g^{-1}). The tunas, tunas-like and other bony fishes presented intermediate Hg concentrations: *E. alletteratus* (763 ± 411 ng g^{-1}), *T. thynnus* (711 ± 303 ng g^{-1}), *T. atlanticus* (626 ± 310 ng g^{-1}), *X. gladius* (575 ± 225 ng g^{-1}), *T. alalunga* (506 ± 402 ng g^{-1}) and *T. obesus* (474 ± 310 ng g^{-1}). Finally, the lowest concentrations were found in *T. albacares* (257 ± 188 ng g^{-1}), *K. pelamis* (164 ± 95 ng g^{-1}) and *C. hippurus* (107 ± 68 ng g^{-1}) (Fig. 2). However, significant differences in Hg concentrations were only observed between groups a and b shown in Fig. 2.

The highest concentrations were reported for *P. glauca* and *I. oxyrinchus*. These species are highly migratory and can travel for thousands of kilometers in the North Atlantic Ocean (Kohler et al., 2002). These characteristics result on high diversity of food items consumed. *I. oxyrinchus* are fast swimmers and are considered the fastest sharks in the world (Compagno, 2001), while *P. glauca* is the pelagic shark species with the highest growth rate (Dulvy et al., 2008). These characteristics support higher bioaccumulation rates and the ability to prey on high trophic level fish. These sharks are top predators, feeding at the highest level of their food chain (Revill et al., 2009; Froese and

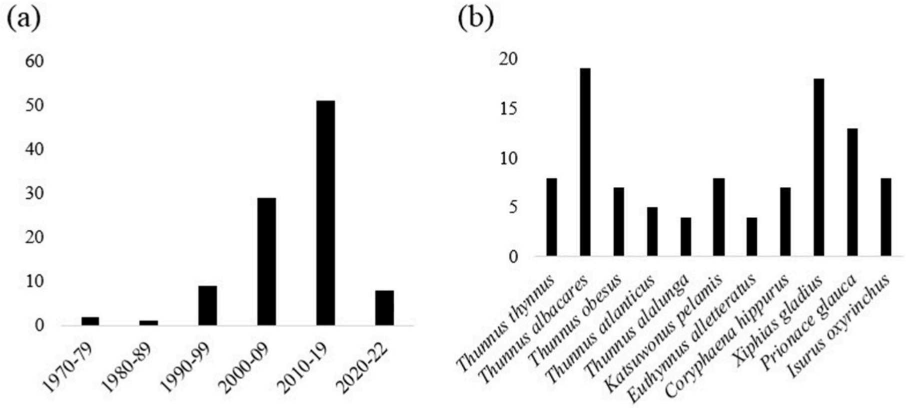

Fig. 1. Distribution of publications in different decades (a) and the number of publications by species (b).

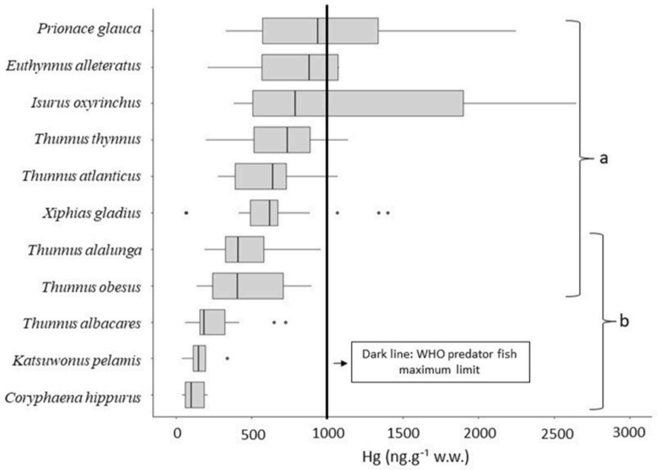

Fig. 2. Boxplot with Hg variations in 11 species of oceanic fish from Atlantic and Mediterranean. Letters a and b represent statistically significant differences.

Pauly, 2022) such cartilaginous fish, marine mammals, and crustaceans, with turtles and seabirds eventually composing their diet (Compagno, 2001). Most specifically, *P. glauca* feeds mainly on teleost, crustaceans and cephalopods (Vaske Júnior et al., 2009), whereas *I. oxyrinchus* feeds mainly on teleost fish and cephalopods, with frequency values higher than 80% frequency for teleost, according to stomach content studies (Maia et al., 2006; Biton-Porsmoguer et al., 2014).

Surprisingly, *E. alleteratus,* a smaller tuna species occupying lower trophic position compared to other bony fish (Kuklyte and Rowe, 2012; Froese and Pauly, 2022), presented the highest Hg levels among tuna species. *E. alleteratus* is a piscivorous species, with a feeding preference for fish, making up almost 70% of its diet (Johnson, 1984). Relatively high Hg concentrations have been reported in *T. atlanticus.* This species has a diet composed mainly by fishes, crustaceans, and squids (Collette and Nauen, 1983; Kuklyte and Rowe, 2012), occupying a high trophic level (Froese and Pauly, 2022), and therefore, exposed to elevated Hg levels due to biomagnification. In addition, *T. atlanticus* feeds in the mesopelagic zone and can dive up to 200 m (Fenton et al., 2015). Organisms found in this region can exhibit high concentrations of Hg, primarily methylmercury, due to microbial methylation in the water column (Choy et al., 2009; Le Croizier et al., 2019).

X. gladius is an oceanic species with a globally distribution, found in tropical and temperate waters, occasionally found in cold ocean waters (Amorim and Arfelli, 1984), but generally found in waters with temperatures above 13 °C (Barrett et al., 1998). It is a species that performs vertical migrations during the day (reaching up to 600 m depth), but at night it prefers more superficial regions, being light intensity a determinant factor (Carey and Robison, 1981). According to data from ICCAT (2006), there are two distinct populations of *X. gladius* in the Atlantic Ocean, one observed in the northern region and the other in the southern region. Other studies using genetic tools, verified four distinct populations of this species worldwide (Mediterranean, North Atlantic, South Atlantic, and Indian-Pacific) (Bremer et al., 1996; Chow et al., 1997; Nohara et al., 2003), which might explain the large variability on Hg levels observed in the present study (Fig. 2). Regarding the feeding of *X. gladius,* there seems to be a distinction between animals captured in the Southeast and Northeast of Brazil. Authors characterize the Southeast region as an important feeding area for swordfish because of the large occurrence of cephalopods, especially squid (Haimovici and Alvarez-Perez, 1990; Zavala-Camin, 1987). However, in the Northeast region, there are studies that show fish as a more frequent and abundant item in the diet of *X. gladius*, probably due to differences in prey assemblage for the Equatorial region (Vaske-Júnior and Lessa, 2005).

The albacore *T. alalunga* is a species that migrates widely for feeding and breeding (Travassos, 1999). Adult specimens prefer temperatures between 10° and 20 °C (Graham and Dickson, 1981), but cooler temperatures (below 9.5 °C) can be tolerated for short time periods (Collette and Nauen, 1983). Oxygen levels and water temperature are dominant factors in the distribution of *T. alalunga* in relation to depth of occurrence, which in the Atlantic is believed to reach 600 m (Collette and Nauen, 1983). In the Atlantic Ocean, *T. alalunga* has a first sexual maturation size between 90 cm and 94 cm, at an age of approximately 5 years (Bard, 1981). Data from ICCAT (2006) identify three distinct populations in the Atlantic Ocean, one in the north, another in the south (separated by latitude 5°N), and one endemic of the Mediterranean Sea. It is one of the main tuna species caught in the South Atlantic Ocean, mainly because it is important for the fish canning companies, which consider such species as a product focus worldwide (Otsu and Uchida, 1959; Collette and Nauen, 1983).

Comparing Hg concentrations between the bluefin tuna, (*T. thynnus*) and the swordfish (*X. gladius*) caught in the Mediterranean, Storelli and Marcotrigiano (2001) found significant differences between these two species, with bluefin tuna showing significantly higher concentrations compared to swordfish, when considering similar weights. The authors concluded that the difference was mainly due to the difference in diets between bluefin tuna (more piscivorous) and swordfish (more cephalopods and molluscs).

Differences in Hg concentrations between tuna species (*T. obesus and T. albacares*) have been reported in various oceanic regions, including the Western Atlantic Ocean, where *T. obesus* shows higher Hg contents than *T. albacares* (Lacerda et al., 2017). Peterson et al. (1973) comparing 88 individuals of *T. albacares* with only 5 of *T. obesus*, also found higher Hg content in the latter species. Differences in diet, behavior, occurrence and foraging depth between the two species may explain their different Hg concentrations. *T. albacares* are migratory, swim near the surface and feed on fish, especially flying fish, squid and pelagic crustaceans, whereas *T. obesus* is also a highly migratory species that feeds in deeper waters (up to 800 m), especially during the day, including demersal carnivorous fish, crustaceans, and squid (Vaske Júnior et al., 2003; Vaske et al., 2012; Silva et al., 2019; Mesquita et al., 2021).

The relatively lower Hg concentrations found in *K. pelamis* and *E. alleteratus* are likely due to their distribution and diet, as these species do not tolerate low oxygen concentrations and temperatures, living most of their lives in shallow waters above the thermocline and consuming smaller prey with lower Hg concentrations (Collette and Nauen, 1983, Choy et al., 2009), when compared to other tuna species (Adams, 2004, Mesquita et al., 2021). Stomach content combined with stable isotope analysis from *K. pelamis* caught in the southwestern Atlantic revealed a feeding composed by lantern fish, krill, and small pelagic fishes, presenting also ontogenetic changes along its life cycle (Coletto et al., 2021). The Hg concentrations in *C. hippurus* were also considered low mostly due to their biology (Adams, 2004, 2009; Cai et al., 2006; Kuklyte and Rowe, 2012), characterized by rapid growth and short life span (less than 2 years), which results in less time to bioaccumulate Hg compared to other species (Adams, 2009). The low trophic level occupied by this species, indicated by stable isotope ratios ($\delta^{15}N$) (Cai et al., 2007; Froese and Pauly, 2022), relative to other species (e.g., *E. alletteratus*) may also be related to the relatively low concentrations observed in *C. hippurus*. Based on stomach content analysis from *C. hippurus* caught in Mediteranean waters, Massutí et al. (1998) reported a diet based mainly on fishes, squids, and crustaceans. Otherwise, Vaske-Jr and Lessa (2004) observed the same pattern in the southwestern Atlantic.

Variations of Hg concentrations as a function of individual size and sex of animals are reported in some of the species studied. For example, the large variation in Hg concentration in similarly sized individuals of *T. albacares* may be due to different growth rates between sexes (Peterson et al., 1973). However, Bosch et al. (2016) found no difference in Hg concentration between sexes in *T. albacares* from South Africa and suggested that different growth rates between individuals of the same sex were perhaps more significant. Methyl-Hg represents more than 98% of the total Hg content in the muscle tissue of *T. albacares* (Voegborlo et al., 2006), the relationship between individual size and Hg concentrations in small-sized fish are unclear, suggesting low biomagnification factor, so environmental factors may be responsible for the higher Hg concentrations. A study of *T. albacares* caught in Florida found that larger females, relative to males, had significantly higher Hg concentrations (Adams, 2004). In a pioneering study (Greig and Krzynowek, 1979) of three different tuna species, no correlation between size and Hg concentration was observed in *T. albacares*, while a significant and positive correlation was established in the other two species studied. A clear relationship between size and Hg content is rarely reported for this species. Nevertheless (Besada et al., 2006), working only with larger individuals (>95 cm) found a significant positive correlation between size and Hg concentrations. As for *T. obesus*, most previous studies on this species, regardless of region, showed a significant relationship between Hg concentrations and body weight (Besada et al., 2006; Choy et al., 2009; Torres et al., 2016). Adams (2004) found significantly higher Hg concentrations in females than in males of *T albacares*, but the females were significantly larger in size. The same author did not find significant differences in Hg concentrations between females and males of *T. atlanticus* and *E. alleteratus*, even though the males were significantly larger in size than the females.

3.2. Geographical differences in Hg concentrations

No significant differences were observed between the different regions when comparing the overall Hg concentrations (including all species) (Kruskal-Wallis chi-squared = 3.0858, df = 3, p-value = 0.3786). Fig. 3 presents a boxplot of the Hg variation among the four oceanic regions studied for the respective species.

Different Hg concentrations in different regions is an issue under discussion. Greig and Krzynowek (1979) for example, found no difference in Hg concentration in *T. albacares* caught in different regions of the Atlantic and Pacific oceans. A study conducted with *T. albacares* associated 12 different capture areas with Hg levels, in this work the authors did not identify significant differences between 3 different regions (NW, NE and SE) of the Atlantic Ocean, but the Northwest Atlantic region showed the highest concentrations (Nicklisch et al., 2017). In this review we compiled data from 18 studies with *T. albacares* in different regions of the Atlantic and found no significant differences between the regions studied, corroborating that previous study.

Concentrations of Hg in *X. gladius* caught in the Atlantic Ocean showed differences within two capture areas, the Southeast region (FAO 27) and the Northeast region (FAO 47) with higher concentrations in the Southeast. However, these differences were not statistically significant ($p > 0.05$) (Esposito et al., 2018). Branco et al. (2007) studying *P. glauca* and *X. gladius* caught in the Equatorial Atlantic and the North Atlantic, found significant differences of Hg concentrations in individuals from these areas, with higher Hg concentrations observed in the Equatorial Atlantic compared to the Azores, in the North Atlantic. On the other hand, Damiano et al. (2011) found no differences of Hg concentrations in *X. gladius* in different regions of the Atlantic. However, significant differences were found for this species caught off the northern coast of Brazil that showed higher concentrations than fish caught off the southern coast (Rodrigues et al., 2013). The authors suggest that this difference may be different Hg loads to those areas, since to more intense gold mining, the largest source of Hg in the region, occurs in the northern coast. (UNEP, 2018). In the first paper that reported Hg data in *P. glauca* in the Atlantic, no significant differences were observed between the Azores and Canary regions (Branco et al., 2004).

The lack of a clear relationship between Hg concentrations in environmental matrixes and the various fish species illustrates the complexity and site-specific nature of Hg bioaccumulation. Thus, direct Hg determinations in the local biota appear to be crucial to adequately evaluating Hg sources, and, ultimately, the risk of the Hg exposure to human health (Castilhos et al., 1998). The absence of significant differences among ocean sub regions is probably due to the origin of most, but not all, of the species included in the reviewed studies, which were obtained by open ocean fisheries. Some, however, included more coastal populations. This difference in sampling area may include areas impacted by local Hg inputs. Along the northeaster continental shelf in Brazil, while sharks caught by open water fisheries show no geographical difference in Hg distribution, more coastal dwelling rays respond clearly to terrestrial sources of Hg (Julio et al., 2022). Therefore, the characteristics of the present review does not allow the establishment of statistically significant differences among ocean sub regions.

3.3. Taxonomical differences in Hg concentrations

According to the reviewed papers, we divided the studied species into different Classes, applying the Kruskal-Wallis test we found significantly higher Hg concentrations in sharks compared to bony fish (Chi-squared = 15.781, df = 1, p-value <0.001) (Fig. 4).

When considering the overall Hg averages (considering all geographic areas), we found a significant higher concentration in Elasmobranchii compared to Osteichthyes (Fig. 4), corroborating previous studies in the Atlantic Ocean. In a study of oceanic fishes in the Northwest Atlantic Ocean, Hg concentrations were found to be highest in mako and thresher sharks, and significantly lower in teleosts (albacore, yellowfin, dolphinfish) (Teffer et al., 2014). In another systematic review with elasmobranchs, it was found that, among different groups, sharks occupying top trophic levels (Carcharhiniformes and Lamniformes) showed the highest Hg concentrations (Tiktak et al., 2020). Higher Hg concentrations in elasmobranchs compared to bony fish, was

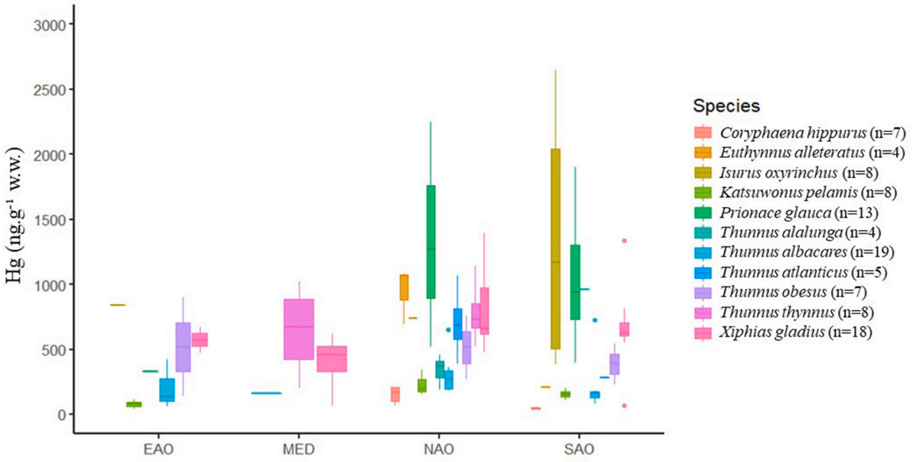

Fig. 3. Boxplot of variation of Hg concentrations in oceanic fish species in the respective capture areas. EAO – Equatorial Atlantic Ocean; MED – Mediterranean; NAO – North Atlantic Ocean; SAO – South Atlantic Ocean.

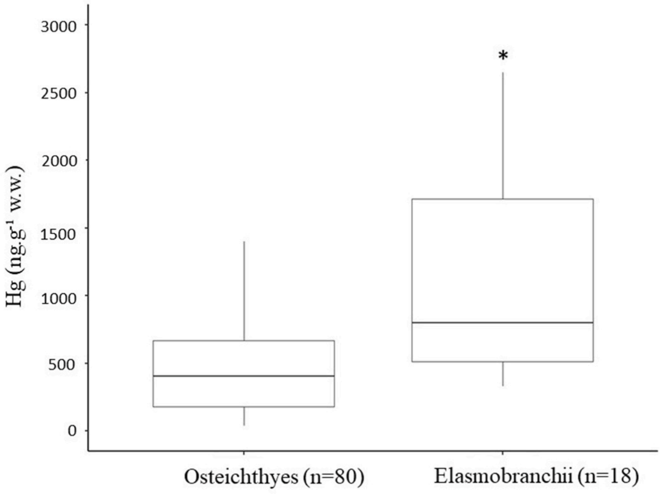

Fig. 4. Boxplot of variation of Hg concentrations in Osteichthyes and Elasmobranchii considering all catch areas. *Statistically significant differences.

also identified in a large systematic review of rays caught worldwide (Bezerra et al., 2019). Even subject to similar diets and habitats, they show higher Hg concentrations than top predatory bony fish, evidencing elasmobranchs have specific characteristics that favors Hg accumulation. Elasmobranchs have slow growth, late maturation, low reproductive rate, longevity, as well as specific metabolic and physiological processes, which make them especially susceptible to contain a high concentration of contaminants and are particularly vulnerable to exposure to pollutants that may pose a risk to the organism (Storelli et al., 2002; Dulvy et al., 2017, 2008; Tiktak et al., 2020). For example, elasmobranchs tend to accumulate more lipophilic compounds in their large livers, which are subsequently deposited in muscle tissues by the joint action of sulfhydryl groups and amino acids (Gelsleichter and Walker, 2010). Sharks also display morphological characteristics such as size and strength of biting, that allow them to prey even on marine mammals and tunas with typically high Hg contents. These differences in elasmobranch physiology and ecology may result in the different concentrations found between Elasmobranchs and Teleostei, however this hypothesis many more studies to be formally tested.

3.4. Human exposure

Data from the Food and Agriculture Organization of the United Nations indicate a world production of 179 million tons of fish, considering fisheries and aquaculture, of which 110 million are destined for human consumption (FAO, 2020). The annual per capita consumption of fish worldwide has increased continuously from 9.0 kg in 1960 to 20.5 kg in 2018 (FAO, 2020). Aquaculture accounted for 46% of the total fish production and 52% of the total fish for human consumption, providing more expansion of fish consumption around the world (FAO, 2020). Global catch includes about 1.700 exploited species, of which finfish account for about 85% of total marine catch production, with small pelagic species being the main group, followed by the order Gadiformes, and tuna-like species. Skipjack (*K. pelamis*) ranked third for the ninth consecutive year with 3.2 million tons. Another species targeted in the FAO Report, that has significant global production is the yellowfin (*T. albacares*), with about 1.5 million tons produced in 2018, ranking 8th overall. Regarding sharks, Brazil, although with a very conflicting fishery management (Neto et al., 2021) is the 11th producer of sharks, the 17th shark fin exporter and first importer of shark meat (Barreto et al., 2017; FAO, 2020). The total blue shark imported into Brazil from several countries is similar to the total national production of sharks and rays combined (approximately 21,000 t) (Barreto et al., 2017). Therefore, the species included in this review contribute significantly to human fish consumption and may pose a measurable human exposure to Hg.

Human food safety advisories relative to the consumption of the seafood species are based on the recommendations that limits Hg levels in fish predators to 1000 ng g^{-1} (FAO/WHO, 2011). Although the overall mean Hg values observed in the present review for all species were below this safety limit, in some of the articles studied it was reported that the mean Hg concentrations for six of the studied species (*P. glauca, E. allteratus, I. oxyrinchus, T. thynnus, T. atlanticus and X. gladius*) exceeded the legal limits, thus making some fish unfit for human consumption (Fig. 2). Another data that is commonly reported by authors in fish and Hg studies is the maximum Hg concentrations found in the species studied. In more than a hundred publications reviewed in this review, only the species *K. pelamis* and *C. hippurus* did not have reported Hg concentrations above 1000 ng g^{-1}. In the Mediterranean, for example, 44% of 169 individuals of *T. thynnus* caught presented Hg concentrations higher than 1000 ng g^{-1} (Storelli and Marcotrigiano, 2001). Besada et al. (2006) reported 16.7% of 30 individuals of *T. obesus* from the North Atlantic with concentrations above that limit. For the shark *P. glauca,* caught in the South Atlantic, 40% of 27 individuals were improper to human consumption (De Carvalho et al., 2014). While 67% of 18 individuals of *X. gladius* caught in the North Atlantic, showed Hg concentrations above the legal limit (Burger and Gochfeld, 2006). From the information in the articles surveyed, sharks (*P. glauca* and *I. oxyrinchus*) had the highest Hg concentrations in all areas. As a case study, if considering only the production imported by Brazil from Uruguay, for example, and defining a theoretical average size of these animals in 30 kg, we have a total of 700,000 units of *P. glauca*, when applying the 40% of unfit for consumption as suggested by De Carvalho et al. (2014), over 280,000 animals were considered contaminated for human ingestion, a good reason why we should pay more attention to the regional consumption of these species.

The level of per capita fish consumption is known to be directly related to the risk of exposure. In Brazil, for example, the national annual average in fish consumption is quite low, around 5–10 kg per capita (FAO, 2020), but in some regions in the Amazon the consumption by riverine communities reaches almost 150 kg per capita per year (Oliveira et al., 2010). Although there are data on average apparent consumption of the population (FAO, 2020), it is still difficult to estimate the individual consumption of a particular person, who may consume much more fish than the regional averages and therefore be more subject to risk. As a suggestion there should at least be warnings about certain species in establishments like fish markets and restaurants, a practice not very common in most countries. Some studies chose to use lower limits (500 or 300 ng g^{-1}) to calculate the percentage of predatory fish samples that had Hg concentrations above the limit. Teffer et al. (2014), used the value set by the US Environmental Protection Agency (USEPA, 2010) and found that 100% of 32 *I. oxyrinchus* individuals and 63% of other predatory fish analyzed in their study exceeded that value (300 ng g^{-1}). A study of *X. gladius* caught in the North Atlantic, using the maximum Hg value of 500 ng g^{-1}, found that 71% of the samples exceeded that limit (Monteiro and Lopes, 1990).

The maximum acceptable daily intake (RfD) of Methyl-Hg has limits, which are 100 ng per day for each kg body weight and restricted to 50 ng per kg body weight for pregnant women (fetuses are more sensitive to Hg toxicity), for women who are breastfeeding and children under 10 years of age (USEPA, 2001). In a study with a real exposure value of the Italian population consuming *X. gladius* and assessing the health risk associated this consumption, Hg intake values were calculated and compared with the provisional tolerable daily intake (PTDI) (0.57 mg/kg body weight) as set by the Food and Agriculture Organization/World Health Organization (FAO/WHO). The data were compared with the PTDI 0.57 mg/kg bw/day set by the European Commission and the results showed a more relevant exposure of children (0.97 mg/kg bw/day), due to their body weight (23.1 kg body weight) than of adults (0.40 mg/kg bw/day) (70 kg body weight) (EFSA, 2012a, b; Esposito et al., 2018). Adverse effects on fetal neurodevelopment may be associated with exposure of mothers, so pregnant women should avoid large predatory fish (Esposito et al., 2018). Therefore, depending on the level of individual consumption concentrations below 1000 ng g^{-1} could possibly be harmful.

4. Conclusion

Large pelagic fishes have been proposed as sentinel species to monitor the presence of persistent contaminants in the oceans, such as Hg, on a regional and even a global scale. These are generally long-lived animals, highly migratory, and occupy upper trophic positions in food webs, which explain their relatively high Hg levels. The present study reviews Hg concentrations in several species of tuna, tuna-like and sharks caught in the Atlantic Ocean and the Mediterranean Sea, and identified differences between sharks and bony fish, with significantly higher Hg concentrations in sharks, but no significant difference evidenced among the four ocean sub regions evaluated, regardless of species.

Perhaps it is time to make a criticism of the values established (predator - non-predator) by the main health regulatory agencies in the world, since they establish higher values precisely for species that have high levels of methylmercury (most toxic organic form), reaching in some cases 95% of the total Hg. We must consider that the world fish trade has a high level of disorganization, lack of traceability of the production chain, and sometimes it is difficult to identify the species consumed, especially sharks that are subject to mislabeling on packaging, so that it is almost never indicated which species are part of the final product, which is an additional difficulty in prevention measures. Children and adults who consume sharks once a week are exposed to a higher amount of Hg than recommended by the US EPA, which may pose a risk to these consumers. In other words, species with the characteristics of those studied in this study have a higher contaminant potential and therefore should have a lower maximum allowable limit than the one currently set.

We know that the per capita consumption level of these fish is directly related to the risk of exposure, but there are some reports in the literature that seem to relate to our criticism. For, in some of the papers reviewed, the authors chose to use lower limits to calculate the percentage of samples that had Hg concentrations above the limit, as can be seen in our supplemental material. It is important to emphasize that fish consumption is extremely beneficial for humans, as they are part of a balanced diet and provide large amounts of Omega-3. However, a detailed monitoring of Hg concentrations in individuals of different sizes of target species is needed. The identification of geographical differences in Hg concentrations and the mechanisms behind these trends should help authorities to improve fish consumption advisories. Raising public awareness of Hg exposure risks is paramount, particularly for risk groups, such as pregnant and lactating women and children.

Credit author statement

Felipe A. de Alencar Goyanna: Conceptualization, Methodology, Data curation, Writing- Original draft preparation, Visualization and Investigation. **Moises Fernandes Bezerra**: Data curation, Software, Writing- Reviewing and Editing. **Guelson Batista da Silva**: Writing-Reviewing and Editing. **Luiz Drude de Lacerda:Supervision,** Validation, Writing- Reviewing and Editing.

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Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

Data will be made available on request.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.

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