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Ecological drivers of mercury accumulation in oceanic apex predators: A human consumption advisory

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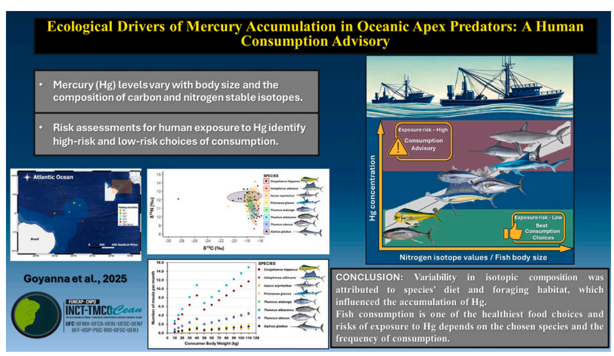
HIGHLIGHTS

- Mercury concentrations vary among various oceanic apex predators.
- Oceanic apex predators present wide variation of isotopic niche ($\delta^{13}C$ and $\delta^{15}N$).
- Shark species accumulate the greatest concentrations of mercury.
- Exposure estimates and fish consumption advisories are provided for largely commercialized oceanic species.

GRAPHICAL ABSTRACT



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ABSTRACT

This article provides a detailed assessment of mercury (Hg) concentrations and stable isotopes ($\delta^{15}N$ & $\delta^{13}C$) in sharks and pelagic fishes of high trophic level caught in the Western Equatorial Atlantic Ocean and presents advisories for safe consumption by human populations. We found significant differences in Hg concentrations among species, with highest Hg concentrations in *Isurus oxyrinchus* (2173.6 ± 1521.5 ng g^{-1}) followed by *Xiphias gladius* (1600.3 ± 671.9 ng g^{-1}) and *Prionace glauca* (1293.9 ± 830.8 ng g^{-1}), while lowest Hg concentrations were observed in *Thunnus albacares* (170.0 ± 42.6 ng g^{-1}) and *Coryphaena hippurus* (190.7 ± 82.8 ng g^{-1}). Intermediate Hg concentrations were observed in *T. alalunga* (512.9 ± 145.9 ng g^{-1}), *Istiophorus albicans* (493.1 ± 348.6 ng g^{-1}) and *T. obesus* (327.7 ± 343.1 ng g^{-1}). The methylmercury proportion relative to total Hg was higher than 80 % for the species *C. hippurus*, *P. glauca*, *T. alalunga* and *X. gladius*. The $\delta^{15}N$ values corroborate with all species occupying upper trophic levels and ranging from 10.2 ± 0.4 ‰ in *T. albacares* to 12.4 ± 1.1 ‰ in *X. gladius*. The highest values were observed in *I. oxyrinchus*, *X. gladius* and *P. glauca*, compared to *C. hippurus*,

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I. albicans and *T. albacares*, which showed the lowest $\delta^{15}N$ values. The $\delta^{13}C$ values ranged from −18.4 ± 2.6 ‰ in *X. gladius* to −16.6 ± 0.4 ‰ in *C. hippurus*, with significant differences between the species. We found significant correlations between Hg concentrations and fish weight for all species, except *C. hippurus*, *I. albicans* and *I. oxyrinchus,* indicating the process of bioaccumulation. The significant and positive correlation between log-transformed Hg concentrations and $\delta^{15}N$ indicates biomagnification in the upper trophic levels of this oceanic food-web. The estimated species-specific number of meals that are safe for consumption ranged from 0 to 11 meals per month for adults and children.

1. Introduction

Mercury (Hg) is an environmental concern that can negatively impact the health of humans and wildlife (Mills et al., 2024). In pelagic oceanic areas, the main source of Hg is atmospheric deposition, predominantly in the form of Hg^{2+} that undergo biotic and abiotic methylation, in the photic zone, forming methylmercury (MeHg) (Obrist et al., 2018). The methylmercury present in the marine ecosystem can be assimilated in the food web, through direct uptake from the water by phytoplankton, and be transferred to other organisms through feeding (Gosnell et al., 2021). The Hg accumulation in fish is linked to their body size, age, habitat, trophic position, and lifespan, while distribution among tissues and organs is associated to physiological characteristics (Schneider et al., 2013). Biomagnification of Hg in aquatic food webs results in upper-level predators accumulating this pollutant in concentrations several times higher than those observed in primary producers (Lavoie et al., 2013). Many upper-level pelagic fish can reflect spatial variations in Hg contamination across multiple oceanic regions, which can help understanding Hg environmental fate in the marine ecosystem (Goyanna et al., 2023) and highlighting the importance of evaluating multiple species in biomonitoring efforts (Médieu et al., 2023).

Long-term Hg monitoring in these fish is limited and current information on Hg accumulation shows greater than expected levels considering changes in global atmospheric emissions and deposition (Médieu et al., 2023). Furthermore, there are significant gaps in Hg data for many pelagic species from oceanic regions in the South Atlantic Ocean (Goyanna et al., 2023). The monitoring of these organisms can provide complementary information on the vertical and horizontal distribution of methylmercury in the ocean (Goyanna et al., 2023; Médieu et al., 2023; Lacerda et al., 2017). However, to use these fishes as biomonitors of Hg levels, in accordance with the Minamata Conventions on Mercury, it is necessary to understand ecological drivers of Hg accumulation and to disentangle these from the spatial variability of Hg concentrations in the ocean.

Ecological characteristics are often described as drivers of Hg accumulation in marine megafauna, including species foraging habitat (including depth), feeding ecology and associated ontogenetic shifts, trophic interactions (predation) and food web structure (Barrios-Rodriguez et al., 2024; Moura et al., 2020). These characteristics can be assessed through Stable Isotope Analysis (SIA) of light elements, such as carbon and nitrogen, in tissues of marine organisms (Peterson and Fry, 1987). The ratios of nitrogen ($^{15}N/^{14}N$) and carbon ($^{13}C/^{12}C$), relative to a reference standard (hereafter $\delta^{15}N$ and $\delta^{13}C$, respectively), are commonly used to examine trophic structure (Post et al., 2007; Yeakel et al., 2016). For example, as $\delta^{13}C$ changes discreetly as carbon moves from the base of the food web to upper-level consumers (enrichment of ~0.5 ‰–1 ‰) and reflects the main sources of carbon for a given food web (Bouillon et al., 2011; Peterson and Fry, 1987). On the other hand, the $\delta^{15}N$ is enriched in consumers (~2 ‰–4 ‰) relative to their diet which can provide an estimate of trophic position and characterization of trophic niche (Hussey et al., 2010; Layman et al., 2007; Post, 2002). Therefore, the use of SIA can complement the assessment of Hg accumulation in predator fish. As $\delta^{15}N$ generally increases with higher trophic levels and Hg also accumulates at these levels, a positive relationship can be observed between $\delta^{15}N$ values and Hg concentrations across multiple species in the same food web. This is especially useful for characterizing the biomagnification of Hg in the trophic web, defined as the average increase of Hg concentration as it moves up the food web (Condini et al., 2017; Sinkus et al., 2017).

The production of aquatic organisms globally peaked in 2018 with 178.9 million tons, including wild capture fisheries and aquaculture, and plateaued out through 2019 and 2020 (FAO, 2022). The average production for the last three years of available data is 178 million tons per year, with marine fish harvesting accounting for approximately 46 % (81.1 million tons) of this amount. Pelagic fish species accounts for a major fraction of fisheries production globally, with global catches of tuna and tuna-like species reaching 7.8 million tons in 2020, from which the Skipjack tuna, *Katsuwonus pelamis* and Yellowfin tuna, *Thunnus albacares*, account for 55 % of catches in this group (FAO, 2022). Brazil is an important player in the fishing activity carried out in the Atlantic Ocean, despite having a highly contentious fishery management system (Neto et al., 2021). The total catches reached 52,519 t in 2021, including tuna and tuna-like species, and other large pelagic species (e.g., billfish, sharks, wahoo, dolphinfish, etc.) (ICCAT, 2022). Specifically, total catches per species type in 2021 were 4629 t for the Blue Shark, *Prionace glauca*, 2110 t for the Swordfish, *Xiphias gladius*, 6.499 t for the bigeye tuna, *T. obesus*, 13,664 t for the yellowfin, *T. albacares,* 516 t for the albacore, *T. alalunga,* 477 t for the Mako shortfin, *Isurus Oxyrinchus* and 24 t for the Atlantic sailfish, *Istiophorus albicans* (ICCAT, 2022). Provisional estimates suggest that 88.4 % of the average global production is intended for human consumption, resulting in a per capita apparent consumption of 20.4 kg per year (or 55.9 g per day) (FAO, 2022).

Considering that fish consumption is the main route of Hg chronic exposure in human populations (UNEP, 2018), it is important to estimate human exposure to Hg through consumption of these large pelagic predators Risk assessments often show that factors such as, species type and size, frequency of consumption and Hg levels are the most important factors governing human exposure through fish consumption (Bezerra et al., 2023), but exposure assessments are still limited for these large pelagic predators (Goyanna et al., 2023).

Mercury levels in apex oceanic predators are generally in the organic, and most toxic, form of methylmercury (>80 %) (Monteiro and Lopes, 1990; Storelli and Marcotrigiano, 2001). Fishery activity along the Brazilian coast accounts for <1 % of the global fish catches (FAO, 2022), still MeHg exports through fish catch in this region accounts for 21 % of total. (Wu and Zhang, 2023). These estimates show that upper trophic level fish from the Brazilian Equatorial Atlantic Coast, including the Amazon River discharge area, are among the highest in MeHg concentration globally (Wu and Zhang, 2023).

Therefore, the goals of the present study are three-fold 1) Assess Hg and MeHg accumulation in upper trophic level pelagic fish in the Equatorial Western Atlantic Ocean; 2) Evaluate ecological drivers of Hg accumulation using $\delta^{15}N$ and $\delta^{13}C$; and 3) Estimate human exposure to Hg through fish consumption while providing consumer-friendly recommendations on the best choices for consumption.

Specifically, we ask the following research questions to guide our study: 1) Do Hg accumulation levels in upper trophic level fish vary across species and geographical location? Based on global data on Hg accumulation in pelagic fish species, we hypothesize that Hg levels in upper trophic level pelagic fish from the Equatorial Western Atlantic varies by species and are lower than those in similar species from other oceanic regions reflecting previous measurements of dissolved Hg in oceanic waters (Lamborg et al., 2014). 2) Do species-specific ecological characteristics (e.g. isotopic niche) explain Hg accumulation in upper trophic level pelagic fish? Given that $\delta^{15}N$ and $\delta^{13}C$ can provide useful information on foraging habitat, diet, and trophic level, we hypothesize that isotopic niche and other ecological characteristics partly explain variability in Hg accumulation among species. 3) Do upper trophic level pelagic fish accumulate Hg at levels potentially harmful to human consumers? Given the high trophic position of these fish and the biomagnification of Hg in marine food webs, we hypothesize that Hg levels in these large pelagic fish are potentially harmful to human consumers. Therefore, advisories on the frequency and amount of consumption might be necessary.

2. Material and methods

2.1. Sampling description

Muscle samples of *Coryphaena hippurus* ($n = 17$)*, I. albicans* ($n = 8$)*, I. oxyrinchus* ($n = 7$)*, P. glauca* ($n = 50$)*, T. alalunga* ($n = 44$)*, and X. gladius* ($n = 44$) were obtained directly from the fishing industry, during landings of licensed vessels operating in the Northeastern coast of Brazil during two seasons (August 2020 and October 2022). The muscle samples of *T. obesus* ($n = 30$) and *T. albacares* ($n = 52$) were obtained directly from the tuna fleet, during two distinct seasons (July 2014 and October 2015), operating around the PIRATA Program (Pilot Moored Array in the Tropical Atlantic) buoy anchored at position 0° N; 35° W. Employed capture techniques included, trolling, hand lines (HL), and longlines (LL) (Silva et al., 2016). All species were caught between 3°N - 6°S and 16° - 30° W geographical coordinates (Fig. 1).

Fish individuals were weighed (in kilograms), and a ~50 g sample of caudal muscle tissue were collected, avoiding fillets. Eviscerated individuals or those showing signs of previous treatment for conservation were avoided. Samples were immediately placed in sterilized plastic bags and frozen at −20 °C until processing in the lab. Sample processing included freeze-drying for 48 h to 96 h, followed by manual homogenization using mortar and pestle until a fine powder was obtained. All instruments and utensils used for sample processing were previously cleaned in an acid bath (HNO_3 10 %) and ultrapure water.

2.2. Mercury and methylmercury analysis

The quantification of total Hg (THg) in the samples was carried out using a NIPPON NIC RA-3 Cold Vapor Atomic Absorption Spectrophotometer (CV-ASS). Muscle samples weighing approximately 0.5 g were placed in Teflon tubes with 10 mL of concentrated nitric acid (HNO_3 65 %. Tubes were then placed in a microwave oven with 1600 W power at 200 °C for 30 min. After cooling down, 1 mL of hydrogen peroxide (H_2O_2) was added to avoid re-complexation of Hg to the organic matter. The final extract solution was diluted to 100 mL in volumetric flasks with ultra-pure water. For each analysis, the materials used were washed in a bath of neutral detergent followed by HCl 10 %. The calibration curve was constructed using a stock Hg solution (1000 μg mL^{-1} MERCK) with concentrations varying from 1 ng mL^{-1} to 5 ng mL^{-1}. For the quantification of methylmercury (MeHg), about 100 mg (wet weight) of each muscle tissue sample with 5.0 mL of KOH: methanol solution (25 % *w/v*, Merck) was added to extract MeHg in an oven (Nova Instruments, Model NI 1512, Brazil) at 70 °C for 6 h, with gentle stirring applied every hour. The samples were then kept in the dark to avoid possible degradation of MeHg. As described by Taylor et al. (2011), ethylation was carried out with 200 μL of $C_2H_3NaO_2$ buffer (2.0 mol L^{-1}, pH 4.5) followed by the addition of 30 μL of sample and 50 μL of $NaBr_4$ solution (1 % w/v, Brooks Rand Labs, Seattle, USA). The final volume of 40 mL was attained with ultrapure water (Milli-Q, Milli-pore, USA). The MeHg content was then determined with a gas chromatograph coupled to an atomic fluorescence spectrophotometer (Merx™, Brooks Rand Labs, Seattle, USA). Certified reference materials (ERMBB422: 601 ± 30 ng g^{-1} of THg; and BCR-463: 304 ± 16 ng g^{-1} of MeHg) were digested and analyzed, in duplicate, for every sample batch ($n = 26$) with average recoveries of 109.8 % and 87.2 % for THg and MeHg, respectively.

2.3. Stable isotope analysis

Dry muscle samples weighing about 0.5 mg (dry weight) were placed in tin capsules followed by elemental and isotope ratios of carbon ($^{13}C/^{12}C$) and nitrogen ($^{15}N/^{14}N$) quantification in an isotopic mass spectrometer (Thermo Finnigan Delta V Advantage) coupled to an organic elemental analyzer (Flash 2000 - Thermo Fisher Scientific). Analytical control was performed for every 10 samples using certified isotopic standards (Elemental Microanalysis Protein Standard OAS: 46.5

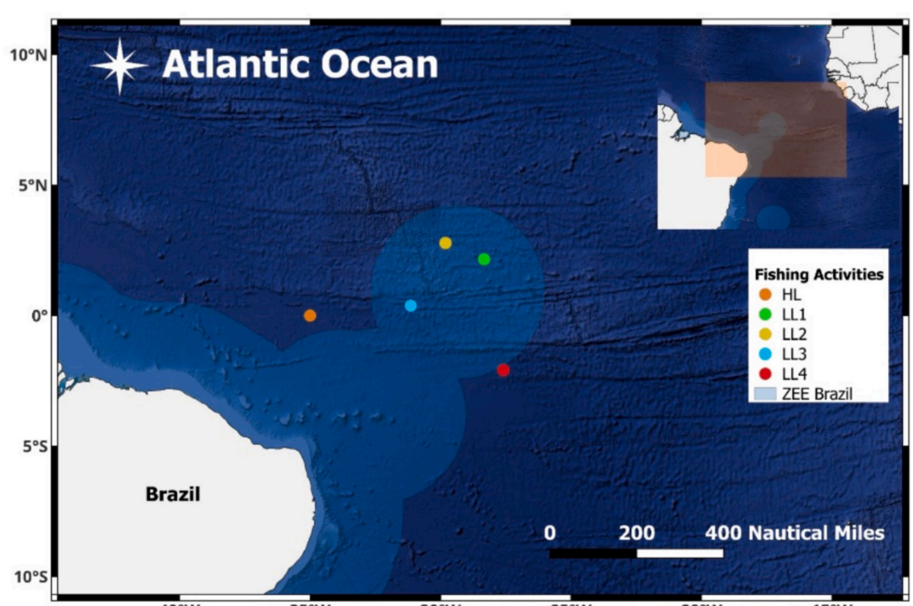

Fig. 1. Location of the study area in the Western Tropical Atlantic Ocean. Colored dots in the inset map depict the location of fishing activities employing the pelagic longline (LL), and handline (HL) fishing methods. The light blue shading represents the Brazilian exclusive economic zone (ZEE).

± 0.78 ‰ for C; 13.32 ± 0.40 ‰ for N; − 26.98 ± 0.13 ‰ for δ^{13}C; + 5.94 ± 0.08 ‰ for δ^{15}N). Reproducibility and accuracy of 95 % was measured using replicates ($n = 3$) and a certified standard (Protein cat. B2155). Results are expressed in the conventional delta (δ) notation relative to Pee Dee Belemnite for δ^{13}C and atmospheric N_2 for δ^{15}N, with precision of 0.1 ‰, and the detection limits for C and N were 0.05 ‰ and 0.02 ‰, respectively.

2.4. Human exposure risk assessment

Following recommendations of USEPA (2000, 2001 and 2022) for Hg exposure assessments and protocol described by Bezerra et al. (2023) we estimated the following parameters. The fish safety level (FSL), defined as the reference Hg concentrations (mg kg^{-1} wet weight) in edible fish tissues that pose no potential risk to fish consumers (USEPA, 2001). This parameter was determined using Eq. (1).

$$FSL_{local} = \frac{BW \times RfD}{CR_{local}} \tag{1}$$

where BW is the average consumer body weight in kilograms, CR_{local} is the daily fish consumption rate (in kg day^{-1}), and RfD is the reference dose of Hg daily ingestion (0.0001 mg kg_{BW}^{-1} day^{-1}). We estimated FSL parameter for two fish consumption scenarios, 1) A CR_{local} of 0.0245 kg day^{-1} reflecting the Brazilian per capita fish consumption for northeastern Brazilian population (e.g. general) (IBGE, 2021), and 2) A CR_{local} of 0.142 kg day^{-1} reflecting fish intake of populations that eat fish daily or as subsistence. We also estimated Hg exposure for two consumer populations, 1) Adults over 18 years old (70 kg of body weight) and 2) Children under 6 years old (15 kg of body weight).

The maximum monthly allowance of meals (CR_{max}) for each fish species was calculated by modifying Eq. (1), as shown below:

$$CR_{max} = \frac{BW \times RfD}{C_{fish}} \tag{1.2}$$

In this modified equation, C_{fish} (mg kg^{-1}) represents the Hg concentration quantified in the studied fish. Subsequently, we estimated the safe number of meals per month (CR_{mm}) using Eq. (2):

$$CR_{mm} = \frac{CR_{max} \times T_{ap}}{MS} \tag{2}$$

where T_{ap} is the averaging period (365.25 days per 12 months or 30.44 days per month), and MS represents the average meal size (in kg meal^{-1}) (USEPA, 2000). For our calculations, we assumed an average fish meal size of 0.15 kg meal^{-1} for adults (BRASIL, 2014) and 0.075 kg meal^{-1} for children. We estimated the daily intake (EDI_{Hg}, mg kg_{BW}^{-1} day^{-1}) for both consumption scenarios and all consumer groups using Eq. (3)

$$EDI_{Hg} = \frac{C_{fish} \times CR_{local}}{BW} \tag{3}$$

Finally, we evaluated the target hazard quotient (THQ) using Eq. (4) according to USEPA (2022). THQ reflects a lifetime chronic non-carcinogenic exposure risk from ingesting seafood-derived Hg. A THQ below 1 indicates no expected negative effects on health, while a THQ exceeding 1 indicates that Hg exposure may potentially cause adverse health effects.

$$THQ = \frac{EF \times CR_{local} \times ED \times C_{fish}}{RfD \times BW \times AT} \tag{4}$$

where EF is the exposure frequency (365 days/year), ED represents the exposure duration (77 years for adults and 6 years for children), and AT stands for the averaging exposure time (EF × ED).

2.5. Statistical analysis

All statistical analyses and plotting figures were performed using R version 3.4.3 (Core Team, 2023). Parametric assumptions of normality and homoscedasticity for the data were tested using Shapiro Wilks and Levene's tests. We conducted non-parametric Kruskal Wallis tests followed by post hoc pairwise Wilcoxon Rank tests, with Holm corrections, to test for differences in Hg accumulation and δ^{13}C values among species. We conducted parametric ANOVA followed by post-hoc Tukey test to test differences in δ^{15}N among species. We used Pearson's correlation to assess the relationship between THg and fish weight, and Spearman's correlation to check the relationship between THg and other variables. Statistical significance was set at $p = 0.05$ for all tests. Using the Stable Isotope Bayesian Ellipses (SIBER) package (Jackson et al., 2011) we calculated isotopic niche space metrics (e.g., Standard ellipse area corrected for small sample size (SEA_c), Bayesian standard ellipse area (SEA_b), δ^{13}C and δ^{15}N ranges) for each species.

3. Results

We sampled a total of 252 individuals of 8 upper trophic level pelagic species, including six teleost species; the Dolphinfish (*C. hippurus*, $n = 17$), the Atlantic Sailfish (*I. albicans*, $n = 8$), the Albacore tuna (*T. alalunga*, $n = 44$), the Bigeye tuna (*T. obesus*, $n = 30$), the Yellowfin tuna (*T. albacares*, $n = 52$), and the Swordfish (*X. gladius*, n = 44); and also two elasmobranch species, the Shortfin mako shark (*I. oxyrinchus*, $n = 7$), and the Blue shark (*P. glauca*, $n = 50$). We included in our analysis the recently published Hg data for *T. alalunga* reported by Lacerda et al. (2024), as well as those for *T. obesus* and *T. albacares* reported by Lacerda et al. (2017) considering that quantified additional variables for the same individuals, such as δ^{13}C, δ^{15}N, and MeHg.

Overall, the average fish weight varied from 9.7 ± 7.4 kg in *C. hippurus* to 60.9 ± 13.5 kg in *I. oxyrinchus* (Table 1). The smallest individual sampled was a *T. albacares* weighing 1.3 kg, and the largest individual was an *X. gladius* weighing 127.1 kg (Table 1).

3.1. Mercury (Hg) accumulation

Mercury data were converted to a wet weight basis, using moisture content of muscle tissues for the respective fish species (overall average of 75 ± 3.4 %) and expressed in ng g^{-1}. Methylmercury were quantified in a subset of samples from four of the eight species studied. The average proportion relative to THg was 82.9 ± 5.3 % in *C. hippurus* ($n = 2$), 82.0 ± 21.4 % in *P. glauca* ($n = 29$), 92.0 ± 15.2 % in *T. alalunga* (n = 3), and 82.1 ± 1.9 % in *X. gladius* ($n = 16$). Overall, considering these four species, an average of 84.8 ± 4.2 % of THg is organic MeHg.

Median THg concentrations were highest in *I. oxyrinchus* (2152.8 ± 1110.9 ng g^{-1}), followed by *X. gladius* (1414.6 ± 595.1 ng g^{-1}), and *P. glauca* (998.6 ± 666.9 ng g^{-1}). Median THg concentrations were lowest in *T. albacares* (136.5 ± 85.8 ng g^{-1}) followed by *C. hippurus* (157.7 ± 78.3 ng g^{-1}). Intermediate median THg concentrations were observed in *T. alalunga* (488.9 ± 217.4 ng g^{-1}), *I. albicans* (487.2 ± 484.7 ng g^{-1}), and *T. obesus* (432.3 ± 451.1 ng g^{-1}) (Table 1). The lowest and highest THg concentration was observed in *I. albicans* (75.3 ng g^{-1}) and *I. oxyrinchus* (5159.6 ng g^{-1}) individuals, respectively (Table 1). We found significant differences in THg concentrations among species (Kruskal-Wallis H = 197.0; df = 7; $n = 252$; $p < 0.001$) (Fig. 2).

3.2. Stable isotope values (δ^{15}N and δ^{13}C) and isotopic niches

We determined stable isotope ratios for a subset of sampled individuals totaling 133 individuals, including *C. hippurus* (17), *I. albicans* (8), *I. oxyrinchus* (7), *P. glauca* (29), *T. albacares* (5), *T. alalunga* (42), *T. obesus* (5) and *X. gladius* (20).

The average δ^{15}N values ranged from 10.2 ± 0.4 ‰, in *T. albacares*, to 12.4 ± 1.1 ‰ in *X. gladius* (Table 2). Significant differences among

Table 1
Mean, standard deviation, minimum and maximum values for weight, total mercury (THg) and methylmercury (MeHg) in 8 species of oceanic fish from the Western Equatorial Atlantic Ocean.

Species (n)	Weight (kg)				THg (ng g^{-1} w.w.)				MeHg (ng g^{-1} w.w.)			
	Mean	SD	Min	Max	Mean	SD	Min	Max	Mean	SD	Min	Max
Coryphaena hippurus (17; 2[a])	9.67	7.39	4.07	35.97	190.7	82.8	111.6	407.3	166.1	31.4	134.7	197.5
Istiophorus albicans (8)	26.65	8.70	13.00	38.48	493.1	348.6	75.3	1048.7				
Isurus oxyrinchus (7)	60.90	13.54	40.46	77.00	2173.6	1521.5	593.1	5159.6				
Prionace glauca (50, 29[a])	39.28	11.57	16.80	70.00	1293.9	830.8	453.2	4792.0	1001.1	468.6	240.0	2370.5
Thunnus alalunga (44; 3[a])	28.52	4.50	20.80	44.59	512.9	145.9	294.5	930.1	561.9	155.9	342.6	691.2
Thunnus albacares (52)	16.76	13.41	1.31	60.00	170.0	42.6	125.7	236.4				
Thunnus obesus (30)	13.07	9.03	2.92	51.02	327.7	343.1	95.4	1748.1				
Xiphias gladius (44; 16[a])	55.50	19.11	32.75	127.07	1600.3	671.9	646.8	3521.2	1431.7	695.8	657.1	3606.2

n = number of individuals analyzed.
[a] Denotes the number of individuals analyzed for MeHg.

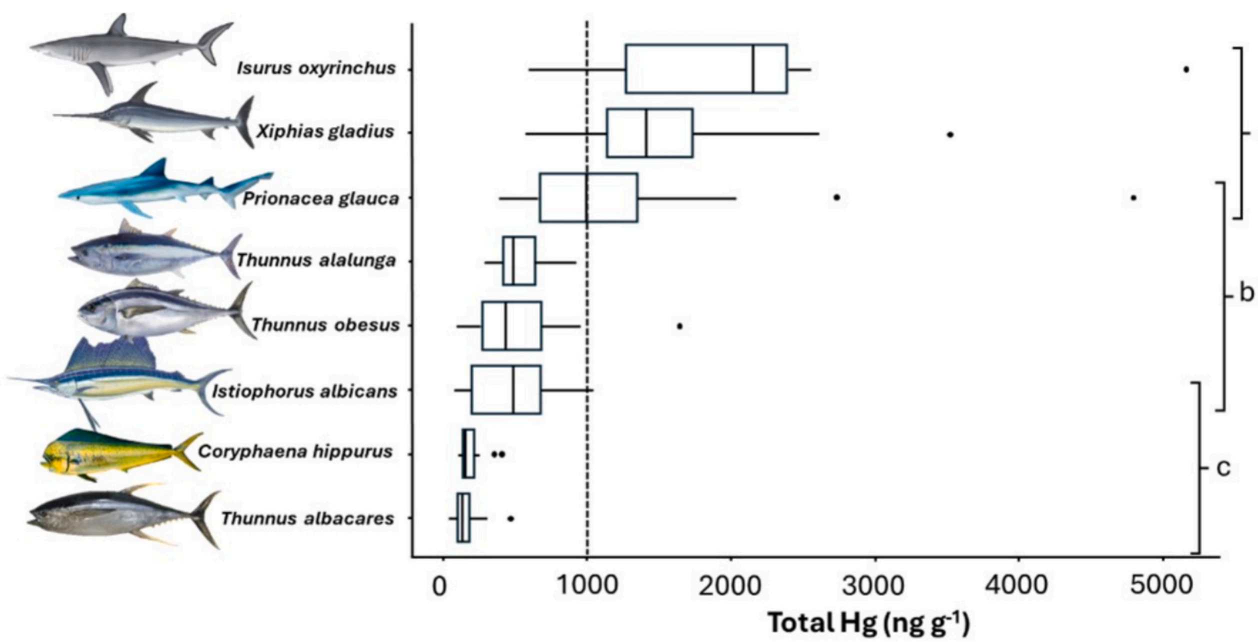

Fig. 2. Total Hg distribution in eight species of oceanic fish from Western Equatorial Atlantic. Letters a, b and c represent statistically significant differences ($p > 0.05$). The dashed line indicates predator fish maximum limit for THg concentration established by the Brazilian regulatory agency (ANVISA, 1998).

Table 2
Descriptive table with the mean, standard deviation, minimum and maximum values for weight, $\delta^{15}N$ and $\delta^{13}C$ of the species studied.

Species (n)	Weight (kg)				$\delta^{15}N$ (‰)				$\delta^{13}C$ (‰)			
	Mean	SD	Min	Max	Mean	SD	Min	Max	Mean	SD	Min	Max
Coryphaena hippurus (17)	9.67	7.39	4.07	35.97	11.0	0.8	9.5	12.0	−16.6	0.4	−17.3	−15.8
Istiophorus albicans (8)	26.65	8.70	13.00	38.48	11.0	15.1	8.6	12.7	−17.4	0.5	−18.2	−16.7
Isurus oxyrinchus (7)	60.90	13.54	40.46	77.00	12.0	10.7	11.0	14.2	−16.8	0.7	−17.7	−16.1
Prionace glauca (29)	40.58	13.54	16.80	70.00	11.9	0.7	10.2	13.2	−17.7	0.8	−20.9	−16.2
Thunnus alalunga (42)	28.64	4.54	20.80	44.59	11.3	0.8	9.8	13.3	−17.4	0.3	−18.0	−17.0
Thunnus albacares (5)	19.61	12.22	2.64	32.20	10.2	0.4	9.8	10.8	−17.0	0.4	−17.8	−16.6
Thunnus obesus (5)	9.73	4.04	5.34	15.51	11.5	0.7	10.5	12.0	−17.4	0.4	−18.0	−17.1
Xiphias gladius (20)	58.16	23.26	35.37	127.07	12.3	0.6	11.3	13.6	−18.4	2.6	−28.5	−16.3

species were observed for $\delta^{15}N$ values (ANOVA $F_{7,125} = 10.76$; $p < 0.001$) (Fig. 3B). The pairwise post-hoc Tukey test ($p < 0.01$) showed that the highest values were observed in *I. oxyrinchus*, *X. gladius*, and *P. glauca*, compared to *C. hippurus*, *I. albicans*, *T. albacares* that presented the lowest values of $\delta^{15}N$. Intermediate values were observed in *T. obesus* and *T. alalunga* (Fig. 3B). The $\delta^{13}C$ values ranged from −18.4 ± 2.6 ‰, in *X. gladius*, to −16.6 ± 0.4 ‰ in *C. hippurus* (Table 2). Significant differences among species were observed for $\delta^{13}C$ values (Kruskal-Wallis $H = 44.7$; $df = 7$, $n = 133$, $p < 0.001$) (Fig. 3A). The pairwise post-hoc Wilcoxon test showed that the highest values were observed in *C. hippurus* compared to *T. alalunga* ($p < 0.001$), *P. glauca* ($p < 0.001$), and *X. gladius* ($p = 0.002$), while no significant differences ($p > 0.05$) were observed for the other pairwise comparisons (Fig. 3A).

The Standard Ellipse Area corrected for small sample sizes (SEA_c), representing the isotopic niche area of each species, and the posterior distribution of estimated Bayesian Standard Ellipses Areas (SEA_b), considering the uncertainty around the isotopic niche areas, are shown in Figs. 4 and 5, respectively. The largest value of SEA_c was found for

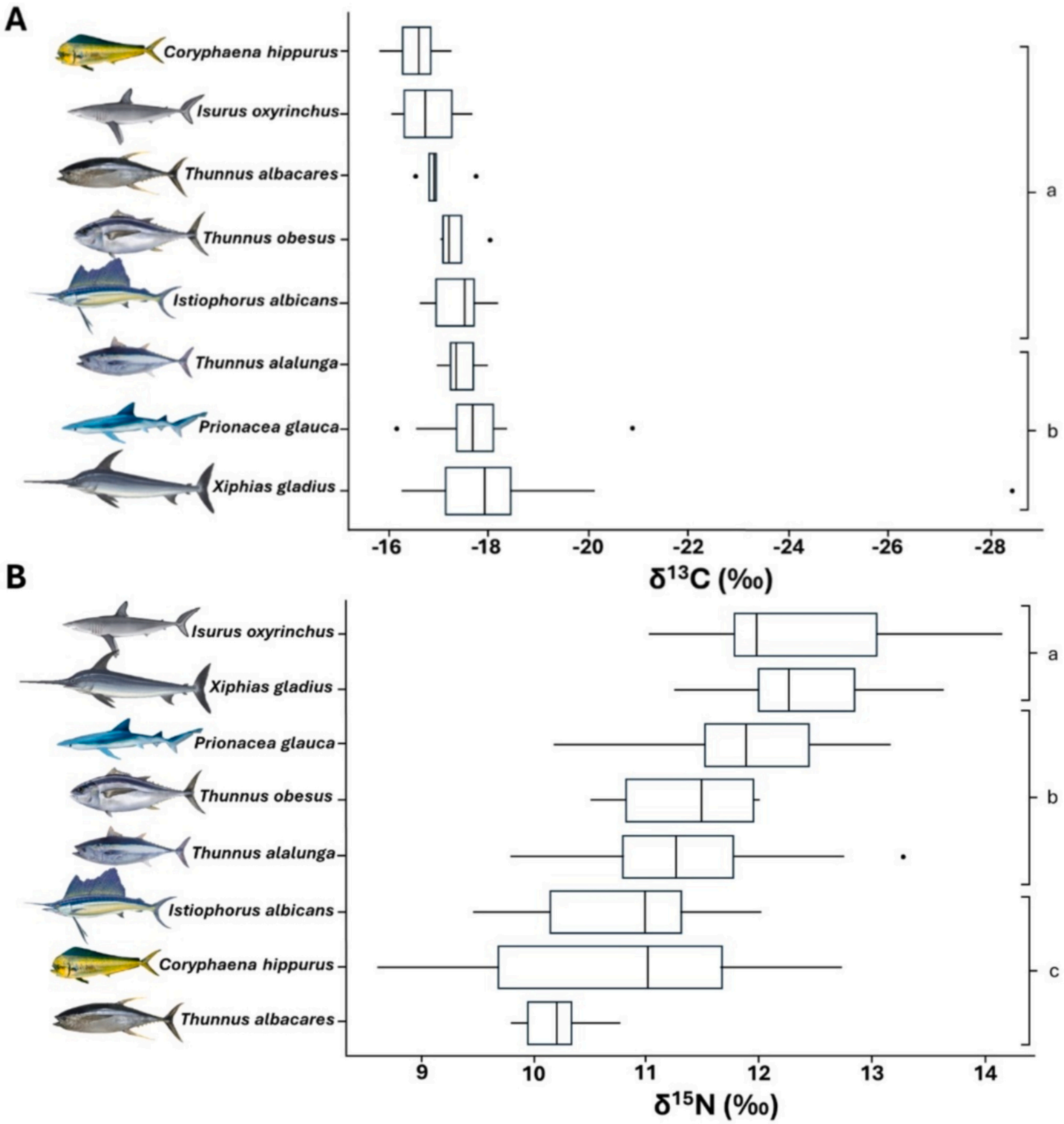

Fig. 3. Distribution of $\delta^{13}C$ (Panel A) and $\delta^{15}N$ Panel (B) values in eight species of oceanic fish from Western Equatorial Atlantic Ocean. Letters a, b and c represent statistical differences ($p > 0.05$).

X. gladius (5.3 $‰^2$), followed by *I. oxyrinchus* (2.6 $‰^2$), *P. glauca* (1.9 $‰^2$), *I. albicans* (1.8 $‰^2$), *C. hippurus* (1.0 $‰^2$), *T. obesus* (0.9 $‰^2$), *T. albacares* (0.7 $‰^2$), and *T. alalunga* (0.6 $‰^2$).

3.3. Correlations among variables

We observed a significant correlation between THg concentrations and fish weight for all species (Fig. 6; Table S1), except *C. hippurus* (Spearman, df = 15, $r = 0.18$, $p = 0.47$), *I. albicans* (Pearson, df = 6, $r = 0.52$, $p = 0.18$), and *I. oxyrinchus* (Pearson, df = 5, $r = 0.25$, $p = 0.58$).

Furthermore, when all species were included, we found a significant and positive relationship between Log-transformed Hg and $\delta^{15}N$ values (Fig. 7). However, when performed on each species separately, correlation test between Log-transformed Hg concentrations and $\delta^{15}N$ was significant for *X. gladius* only (Pearson, $r = 0.54$, df = 18, $p = 0.012$) (Table S1). The correlation test between Log-transformed Hg concentrations and $\delta^{13}C$ was significant for *C. hippurus* only (Pearson, $r = 0.59$, df = 15, p = 0.01) (Table S1).

3.4. Risk exposure estimation and consumption recommendations

We estimated a fish screening level (FSL), for a general intake, of 0.28 mg kg^{-1} and 0.06 mg kg^{-1}, for adults and children respectively. For subsistence intake, we estimated a FSL of 0.05 mg kg^{-1} and 0.01 mg kg^{-1}, for adults and children, respectively. Using mean Hg concentrations for each species, we estimated THQ, EDI_{Hg}, CR_{max}, and CR_{mm}, for

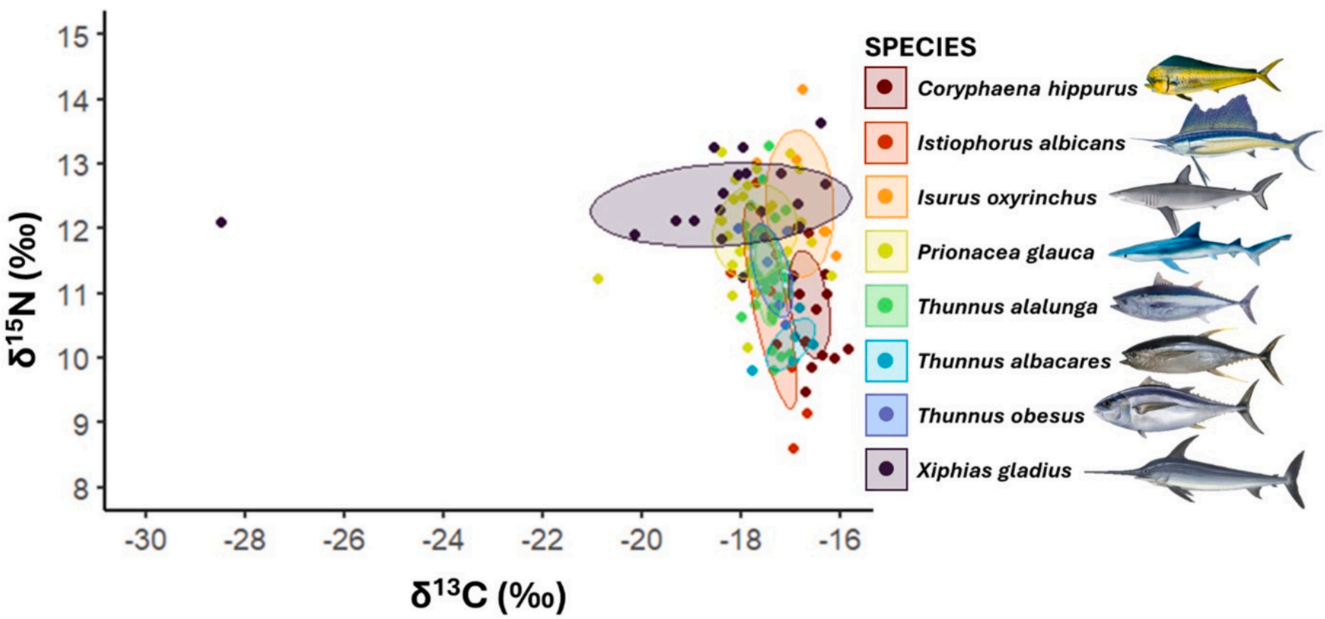

Fig. 4. Isotopic niche of upper trophic level pelagic fish in the Western Equatorial Atlantic Ocean.

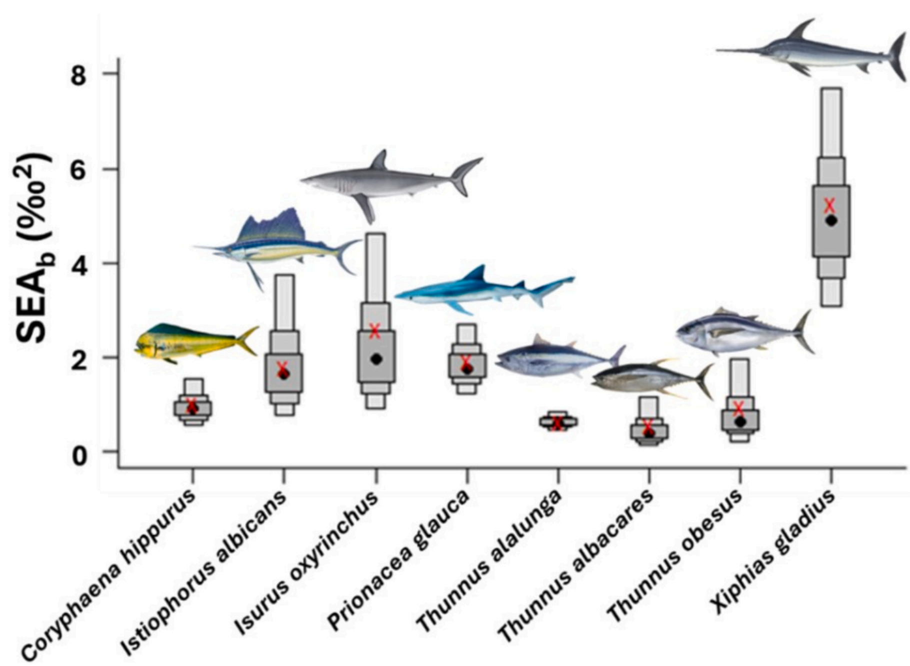

Fig. 5. Bayesian Standard Ellipses Areas (SEA_b) for eight species of oceanic fish from Western Equatorial Atlantic Ocean. The "X" in red represents the estimated Standard Ellipses Areas corrected for small sample sizes (SEA_c).

adults and children under two consumption scenarios, general intake ($CR_{local} = 0.0245$ kg day^{-1}) and subsistence intake ($CR_{local} = 0.142$ kg day^{-1}) (Tables S2 and S3).

We found that THQ values estimated for adult general consumers were above 1 (indicating potentially excessive exposure) for most species, except for *C. hippurus* and *T. albacares*. Similarly, THQ values estimated for adults and children in the subsistence consumption scenarios were above 1 for all species. Considering children consumers, the two scenarios (general and subsistence) showed values above 1 for all studied species. The calculated EDI values varied from 0.0001 ± 0.0000 mg kg_{BW}^{-1} day^{-1} in adults, to 0.0024 ± 0.0009 mg kg_{BW}^{-1} day^{-1} in children in the general consumption scenario. Under the subsistence consumption scenario, the calculated EDI values varied from 0.0004 ± 0.0002 mg kg_{BW}^{-1} day^{-1} in adults, to 0.0024 ± 0.0009 mg kg_{BW}^{-1} day^{-1} (Table S2).

Using mean values of THg concentration, we estimated the maximum consumption rate (CR_{max}) and subsequent number of meals per month (CR_{mm}) for each species (Table S3). The CR_{mm} varied from 0 to 11 meals per month and was highest in *T. albacares* and *C. hippurus*. For species *X. gladius* and *I. oxyrinchus* the estimated safe number of meals was below one meal per month for consumers in the children's group (Fig. 8).

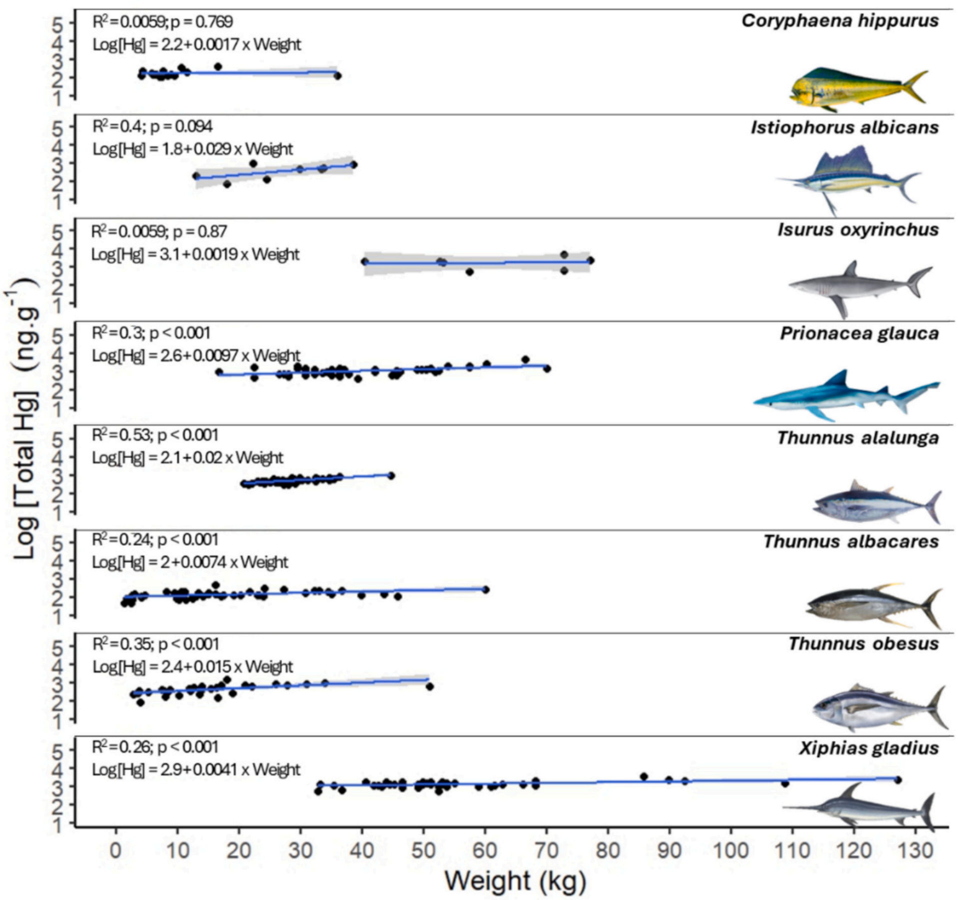

Fig. 6. Relationship between Log-transformed Hg concentrations and fish weight (kg) for eight species of oceanic fish from Western Equatorial Atlantic Ocean.

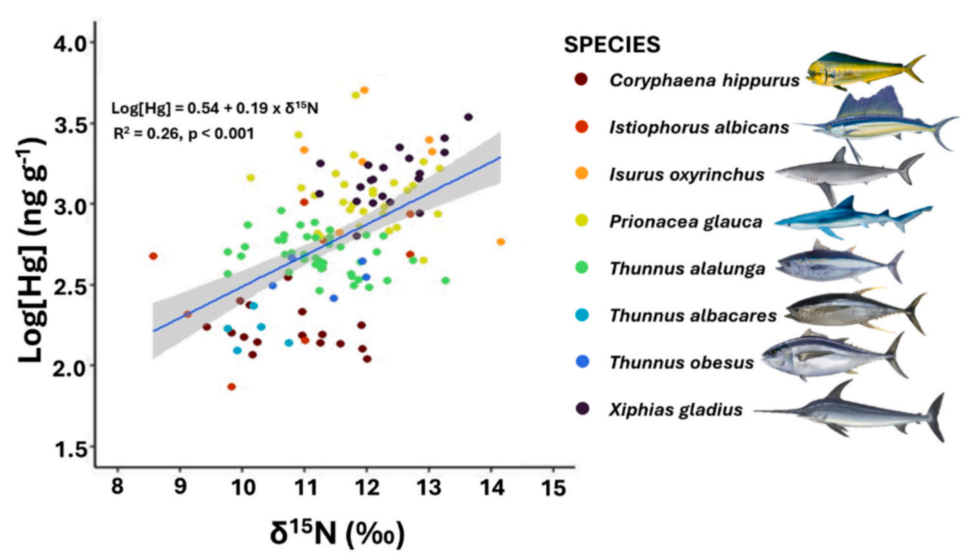

Fig. 7. Relationship between Log-transformed Hg concentrations and $\delta^{15}N$ values in eight species of oceanic fish from Western Equatorial Atlantic Ocean.

4. Discussion

4.1. Mercury accumulation in apex predators of the Western Equatorial Atlantic Ocean

Mercury (Hg) incorporation in oceanic apex predator is primarily governed by diet, which is why foraging ecology largely explains accumulation differences among species (Ferriss and Essington, 2014). However, the emission and deposition processes, and mechanisms regulating MeHg formation in the water column and its assimilation at the base of the food-web determine why species that occur in different geographical areas can present different Hg concentrations in their tissues despite presenting similar diets (Lamborg et al., 2014). Generally, juvenile individuals present lower Hg levels compared to adults,

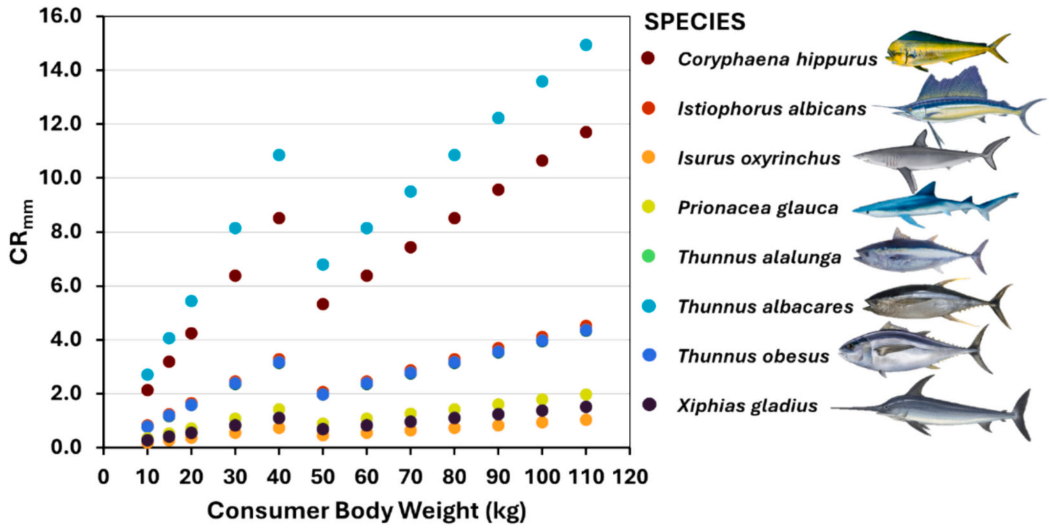

Fig. 8. Variation of recommended fish consumption, in number of meals per month (CR_{mm}), with consumer body weight (kg). Meal size is defined as 150 g and 75 g of raw fish fillet for adults and children, respectively. Children weight varies from 10 to 40 kg, and adults weight varies from 50 to 110 kg.

although this relationship is highly variable among species (Goyanna et al., 2023). The ontogenetic development of endothermic adaptations allows adults to forage at deeper waters where the Hg content of the prey is higher due to intensified methylation processes and the carnivorous nature of deep-water prey (Lamborg et al., 2014; Graham et al., 2007). Furthermore, larger/older individuals present slower growth and lower Hg elimination rates compared to small/young individuals, which result in greater Hg accumulation over time (Ward et al., 2010; Wang and Wang, 2019). Therefore, Hg accumulation in oceanic apex fishes in the present study was closely associated to species-specific life history traits.

In the present study, the largest Hg concentrations were found in two shark species, *I. oxyrinchus* and *P. glauca*, and the bony fish species *X. gladius*. The variability of Hg concentrations between bony and cartilaginous fishes has been previously reported, showing higher concentrations in sharks compared to bony fish of similar habitats (Goyanna et al., 2023). In the present study, elasmobranch species exhibited higher Hg concentrations compared to bony fish despite no clear differention in $\delta^{15}N$ values, indicating that characteristics other than diet such as, slow growth, late maturation, low reproductive rates, and longevity may promote higher Hg accumulation in elasmobranchs (Bezerra et al., 2019; Dulvy et al., 2017, 2008). Previous studies have raised the concern that elasmobranchs can be particularly vulnerable to pollutants and their potential negative health effects (Dulvy et al., 2017, 2008; Storelli et al., 2002; Tiktak et al., 2020; Campos et al., 2024), emphasizing the need for further investigations.

Interspecific Hg concentrations variations can be explained by species-specific life history traits and geographical distribution. The Shortfin mako shark, *I. oxyrinchus,* presented the highest Hg concentrations (2173.6 ± 1521.5 ng g^{-1}) in the present study. This species is considered cosmopolitan with distribution extending tropical and subtropical areas of the Atlantic and Pacific oceans at latitudes between 50° N – 50° S (Compagno, 2005). A previous study in the Southwestern Atlantic (27°08′S-28°38′S and 45°30′W-46°53′W) (Mársico et al., 2007) reported lower Hg levels (384 ± 246 ng g^{-1}) in four individuals of this species. However, because body size of these individuals was not reported, direct comparisons may not be appropriate. In another study, *I. oxyrinchus* individuals captured in the North Atlantic Ocean showed slightly higher Hg concentrations (2647 ng g^{-1}, range 755–4933 ng g^{-1}) in larger individuals (82.6 ± 4.6 kg) compared to those in the present study (Teffer et al., 2014). In contrast, similar Hg concentrations (2110 ng g^{-1}, range 590–5580 ng g^{-1}) were reported for larger individuals (weighing up to 155 kg) in the Eastern South Atlantic Ocean (Watling et al., 1981). In the Pacific Ocean, higher Hg concentrations were found in pectoral and dorsal fins (2500 ng g^{-1} to 5000 ng g^{-1}), a tissue with potentially different accumulation pattern compared to muscle (Kim et al., 2016). Another life history aspect that can influence Hg accumulation is depth distribution, as Hg bioavailability is expected to increase with depth (Médieu et al., 2023). Distribution of *I. oxyrinchus* ranges from 0 to 750 m, but captures are commonly reported between 100 m and 150 m of depth (Weigmann, 2016). Extensive migrations in the Atlantic Ocean can also reflect a wide range of foraging habitats (Kohler et al., 2002), which contributes to the large variation in Hg accumulation in the present study.

The Blue shark, *P. glauca,* is also a highly migratory species, with movement patterns associated with reproductive behavior and prey distribution (Nakano and Stevens, 2008), which contribute to the variability in Hg concentrations across different oceanic areas. The Hg levels observed in the present study corroborate with one previous study in the same area (Carvalho et al., 2014) and other areas in the Pacific Ocean (Escobar-Sanchez et al., 2011; Barrera-García et al., 2012). In contrast, a previous study with individuals from the North Atlantic, found that larger individuals (114.2 ± 11.1 kg) accumulated Hg levels in the same range as those in the present study (Hauser-Davis et al., 2021). This species moves vertically in the water column, with depth distribution of 400 m to 1000 m during the day and up to 150 m at night (Carey et al., 1990; Weigmann, 2016). It is the pelagic shark species with the highest growth rate (Dulvy et al., 2008) and its abundance increases at higher latitudes between latitudes 20° - 40°S (Bigelow et al., 1999; Carvalho et al., 2011).

Among the bony fish species, *X. gladius* showed the highest Hg concentrations. The Hg concentrations found in the present study are the highest average value reported for this species in the Atlantic Ocean, according to a recent review of Hg accumulation in pelagic fishes (Goyanna et al., 2023). Previous studies have reported lower Hg concentrations (1245 ± 729 ng g^{-1}), in larger individuals (103.9 kg, range 564–1485 kg), in the Gulf of Maine (Harding et al., 2018), and in the Mediterranean Sea (490 ± 260 ng g^{-1}), in individuals of similar size (47.2 ± 36.2 kg, range 6–125 kg), compared to the present study (Storelli and Marcotrigiano, 2001). This is an oceanic species, with a global distribution in tropical and temperate waters (Amorim and Arfelli, 1984; Barrett et al., 1998). This species presents vertical migrations during the day, reaching depths of up to 600 m (Carey and Robison, 1981). As mentioned previously, the carnivorous habit of deep-water prey and higher bioavailability of MeHg at deeper water masses can contribute to the observed high Hg levels in this species (Lamborg et al., 2014; Graham et al., 2007).

Moderate Hg concentrations were observed in *T. alalunga, T. obesus* and *I. albicans* in the present study. In general, anthropogenic emissions and methylmercury production in the water column are expected to drive variability in Hg concentrations in these oceanic predators (Médieu et al., 2022).

There is little information about Hg contamination in *T. alalunga* for the Atlantic Ocean as recently shown by Médieu et al. (2023) and Lacerda et al. (2024). Lower Hg concentrations were reported for individuals in the Southeastern Atlantic Ocean (191 ± 87 ng g^{-1}, converted from dry weight basis using 75 % moisture content) (Chouvelon et al., 2017). In contrast, similar Hg concentrations were reported for individuals caught off the southern New England coast (455 ± 144 ng g^{-1}) (Teffer et al., 2014) and in the Gulf of Maine (580 ± 240 ng g^{-1}) (Schartup et al., 2019). Overall, Hg levels in *T. alalunga* for the present study were similar to individuals from the Northwest and Central Pacific Ocean, and from the Indian Ocean, but lower than those from the Mediterranean Sea (Médieu et al., 2023).

The species *T. obesus* is highly migratory, occurs at depths of up to 600 m (Dagorn et al., 2000) and distributes from 50°N – 50°S (Miyake, 1990) in the Atlantic Ocean (Hazin and Travassos, 2007). It feeds in deep waters (400–500 m), consuming carnivorous demersal fish, crustaceans and squids (Mesquita et al., 2021; Silva et al., 2019; Vaske et al., 2012). The Hg concentrations reported for individuals in the North Atlantic were lower than our results, despite their larger body weight (38.9 kg, range 35–43 kg) compared to the individuals in the present study (Yamashita et al., 2005). For the North Atlantic region, Hg concentrations were lower than our results, but for smaller individuals (10.6 ± 0.8 kg) compared to those in the present study (Torres et al., 2016). This species has shown a significant relationship between Hg concentrations and body weight for populations from all oceanic regions (Besada et al., 2006; Choy et al., 2009; Torres et al., 2016). This significant relationship was also observed in the present study.

The species *I. albicans* is a migratory species, which shows rapid growth and voracious carnivorous behavior even during the juvenile stage (Collette et al., 2006). We found only one previous study reporting Hg contamination in *I. albicans* (*I. platypterus* is considered synonymous to *I. albicans* according to Collette et al. (2006)) for the Atlantic Ocean, but in the North Atlantic region (Barber and Whaling, 1983). Direct comparison was not possible as these authors did not report Hg average or range values. For individuals from the Eastern Pacific, Hg concentrations (560 ± 40 ng g^{-1}) were similar to those in the present study, considering individuals of similar body sizes (26.4 ± 0.5 kg) (Bergés-Tiznado et al., 2015). In contrast, higher Hg values (1480 ng g^{-1}) were observed for larger individuals (49–168 kg) in the Southeast Gulf of California (Soto-Jiménez et al., 2010). The diet of *I. albicans* consists mainly of mollusks, especially cephalopods, along with fish from mesopelagic zones (Arizmendi-Rodríguez et al., 2006). This species inhabits areas above the thermocline, typically at depths of 20 m to 40 m, and with temperatures ranging from 25 to 28 °C (Mourato et al., 2014). It is suggested that this species makes seasonal migrations to the Southeast and South regions of Brazil, but there is no certainty about the existence of seasonal variations of *I. albicans* in the Western Equatorial Atlantic Ocean (Hazin et al., 1994; Mourato et al., 2010).

The lowest Hg values were observed in *T. albacares* and *C. hippurus*. Both species are known to feed mainly on epipelagic prey, including fish, squid and crustaceans (Duffy et al., 2017; Brewton et al., 2016; Massutí et al., 1998; Vaske-Jr and Lessa, 2004), which can explain the lower Hg concentrations found in the present study. The species *T. albacares* is found almost all over the world, occurring mainly in tropical and subtropical waters of the Atlantic, Pacific and Indian Oceans (Costa et al., 2005), but not in the Mediterranean Sea (Collette and Nauen, 1983). Mature individuals are distributed in both the surface and deeper areas of the water column (Zavala-Camim, 1987). In general, this species prefers warmer waters compared to other tuna species, and their occurrence increases towards the equatorial region (Hazin, 2006). Previous studies in the same area of the present study, found similar Hg values (Ferreira et al., 2012). Similarly, previous studies from the Gulf of Mexico also reported Hg values in a similar range to the present study (Cai et al., 2007; Senn et al., 2010). In contrast, larger individuals from the latter region reported higher Hg concentrations (Kuklyte and Rowe, 2012), as well as individuals from Northwest Atlantic Ocean (304 ± 87 ng g^{-1}) (Teffer et al., 2014).

C. hippurus makes large feeding and reproductive migrations, with peak catches commonly observed during the summer (Kraul, 1999; Mahon, 1999; Zaouali and Missaoui, 1999). They are generally found at temperatures between 21 and 30 °C and are geographically distributed between 47°N – 40°S, 180°W – 180°L (Palko et al., 1981). *C. hippurus* has been shown to accumulate low concentrations of Hg, probably attributed to its rapid growth and short lifespan (less than two years) (Adams, 2009, 2004). The Hg levels in the present study are similar to those reported for individuals from the Southwest Atlantic Ocean, however those were in smaller individuals (1.9 ± 1.6 kg, range 0.3–6.3 kg) (Moura Reis Manhães et al., 2020). For the Northwest Atlantic Ocean, slightly higher Hg concentrations (205 ± 172 ng g^{-1}) were reported in individuals of smaller weight (4.7 ± 0.4 kg) compared to the present study.

4.2. Stable isotope values in apex predators of the Western Equatorial Atlantic Ocean

The present study is the first report of $\delta^{15}N$ and $\delta^{13}C$ composition in tissues of the studied species for the Western Equatorial Atlantic Ocean. However, previous studies have reported $\delta^{15}N$ and $\delta^{13}C$ values in *C. hippurus*, *T. alalunga*, *T. albacares*, *T. obesus* and *X. gladius* in the Central North Atlantic Ocean (23° N – 43° N) and the Gulf of Mexico (Senn et al., 2010; Logan and Lutcavage, 2013). These are in the same range as those observed in the present study for each respective species. It is worth mentioning that baseline values of $\delta^{15}N$ and $\delta^{13}C$ can vary spatially across oceanic regions and, thus direct comparison might not be adequate. In general, the studied species show $\delta^{15}N$ and $\delta^{13}C$ that are compatible with an oceanic foraging habitat and upper trophic level position across different regions in the South and North Atlantic.

Comparing $\delta^{15}N$ values across the eight studied species, we found a difference of 2.1 ‰ on average between the species with the highest (*I. oxyrinchus*) and lowest (*T. albacares*) $\delta^{15}N$ values. That difference highlights the fact that these are all upper trophic level predators. However, if considering a diet-tissue discrimination factor ($\Delta^{15}N$) of ~2 ‰ as estimated by Hussey et al. (2010) for sharks, it is possible that some species with lower $\delta^{15}N$ values such as, *T. albacares* and *C. hippurus*, can be placed one trophic step lower compared to *I. oxyrinchus*. In fact, both species have been previously identified as prey of this shark species through stomach content analysis (Calle-Morán et al., 2023). Similarly, *X. gladius* and *P. glauca* also presented significantly higher $\delta^{15}N$ values compared to other species. The fact that these three species also presented the highest Hg concentrations supports the notion of these species being at the top of this pelagic food chain. Intraspecific variation in isotope values was considered high for most species. That reflects the variability in size classes for most species studied in the present study. *C. hippurus* presented the broadest range of $\delta^{15}N$ values which can also reflects a generalist foraging behavior and multiple feeding habitats (Brewton et al., 2016). In contrast, the species *T. albacares* presented the lowest range in $\delta^{15}N$ values, which is attributed to the small variability in size classes of sampled individuals. This interpretation is corroborated by the small range in Hg concentrations observed for this species.

Regarding $\delta^{13}C$ ratios, the range values observed for the studied species (−18.0 ‰ to −16.6 ‰) reflects a phytoplankton-based food-web for all these offshore pelagic predators (Troina et al., 2020). Intraspecific variability was highest in *X. gladius, P. glauca* and *I. oxyrinchus*, while *T. alalunga* and *T. obesus* presented the lowest variability. This result reflects the variability in size classes for sharks and the swordfish, *X. gladius,* and suggests these species feed on multiple foraging habitats across areas in the Tropical Atlantic ocean with variable carbon sources

(Graham et al., 2010).

The isotopic niche area models represent the breadth of foraging in oceanic apex predators and are influenced by differences in diet and habitat use of each species, as well as variations in isotopic composition at the base of the food-web (Newsome et al., 2007; Graham et al., 2010). All studied species present a diet composed of a range of prey, including fish, crustaceans and cephalopods. However, the isotopic niches observed in the present study show some species are more specialized than others. The species *T. albacares,* for example, presented the smallest isotopic niche area compared to the other species. Stomach content analysis has shown that this species prefers fish (83.4 %) and shellfish (13.1 %), with a small contribution of crustaceans (1.9 %) (Vaske Júnior et al., 2003). In contrast, the species *I. oxyrinchus* and *X. gladius* presented the widest isotopic niche areas, suggesting a greater variation in foraging habitat and prey choice. Although previous studies have shown these species also feed mainly on teleost fish, reaching 82 % frequency in the cases of bony fish and, to a lesser extent, cephalopods (Biton Porsmoguer et al., 2014), ontogenetic changes in diet can contribute with the observed variation (Kiszka et al., 2015; Chancollon et al., 2006). *I. oxyrinchus* can also occasionally feed on other cartilaginous fish, mammals' marine life, crustaceans, turtles and seabirds (Compagno, 2001). Diet composition of *X. gladius* varies among area of occurrence and includes mesopelagic species, such as myctophids, ommastrephids, and large paralepidids; and epipelagic fish such as Atlantic pomfret, garfish, and Atlantic saury (Chancollon et al., 2006).

Like several species of tuna, *T. obesus* is a highly migratory species that feeds in deep waters (400–500 m), has a very diversified diet, and can be classified as opportunistic, consuming demersal carnivorous fish, crustaceans, and squid (Mesquita et al., 2021; Vaske et al., 2012). The isotopic niche estimated for this species is compatible with its life history traits. Another relevant factor is that several studies suggest ontogenetic dietary changes in tunas (Olson et al., 2016). Graham et al. (2007) observed that juvenile Yellowfin tuna (*T. albacares*) feed mainly on crustacean larvae, while adults consume shrimp and teleost. Young et al. (2010) found an increase in the prey-predator length ratio with an increase in the average length of pelagic predators. These adaptations are reflected in the large intra-specific variability of isotopic values observed in our results. A study formulated a baseline isoscape for the Atlantic Ocean based on a meta-analysis of published plankton $\delta^{13}C$ and $\delta^{15}N$ values (McMahon et al., 2010). Such geographic variations in stable isotope values (or isoscapes) provide a means of tracking the foraging and migration of top marine predators within and between regions at the ocean basin scale. This approach has recently been used to describe the behavior and foraging movements of a wide variety of marine predators, such as tropical tunas (Graham et al., 2010). The adoption of isoscapes helps to explain $\delta^{13}C$ and $\delta^{15}N$ variability within the same species and variability between species. For example, latitudinal increases in muscle $\delta^{15}N$ values for *T. albacares* and *C. hippurus* agree with latitudinal trends for zooplankton (Graham et al., 2010) and likely reflect diminishing influence of N_2 fixation, which is most prevalent in the Caribbean and Sargasso Sea regions (Karl et al., 2002).

Therefore, the variability in the isotopic signature within the same species, or between the species studied, can be explained by the large movement capacity of these animals, considering the way in which they are distributed in tropical areas, they can feed on prey in different trophic chains (value of nitrogen on a different basis) and thus reflect the great variability observed in the studied species, with the exception of *T. albacares* that showed little variability in $\delta^{15}N$ values.

4.3. Risk assessment and intake recommendations

The elasmobranch species, *I. oxyrinchus* and *P. glauca*, exceeded the maximum limit of 300 ng g^{-1} of Hg established by the USEPA (2000), as well as the regional maximum limit of 1000 ng g^{-1} of Hg, established by Brazilian legislation. Among the bony fish species, some individuals of *T. alalunga, I. albicans and T. obesus* exceeded the maximum limit of 300 ng g^{-1} of Hg established by the USEPA, but only *X. gladius* had an average concentration above the maximum limit of 1000 ng g^{-1} of Hg. The *C. hippurus* and *T. albacares* had the lowest Hg concentrations among the others and below the limits set by the USEPA. The fish screening level (FSL) estimated for consumers are concentrations in fish that if exceeded might warrant further investigation as to avoid the risk of excessive exposure to Hg from consuming seafood (Bezerra et al., 2023).

C. hippurus and *T. albacares*, were the only species that did not exceed the calculated FSL levels for adults and children. Fish screening levels are very sensitive to the fish consumption rate, which is why it differs from the commonly used maximum limit of 1.000 ng g^{-1} kg^{-1} established by many governmental agencies worldwide. The EDI and THQ results for the multiple scenarios proposed also indicated a risk for human consuming most species, except *C. hippurus* and *T. albacares.* It should be emphasized that, for the children consumer group, our results were much higher than the safety threshold, indicating the potential for negative effects arising from chronic exposure to Hg through frequent fish consumption. Furthermore, negative effects on fetal neuro development may be associated with exposure of mothers, so pregnant women should avoid these large predatory fish (Esposito et al., 2018).

The annual average fish consumption varies largely by country and is relatively low in Brazil, with about 5–10 kg per capita (FAO, 2020). However, significant variability exists even within a given country as fish consumption has a strong cultural aspect in many human populations and is often consumed on a subsistence basis. For example, in Brazil's Amazonian riverine communities, fish consumption is much higher, with approximately 150 kg per capita annually (Oliveira et al., 2010). Therefore, reporting exposure estimates and parameters such as FSL, EDI and THQ alone are not enough to properly inform consumers of potential risks. Fish consumption advisories should provide objective recommendations, so consumers have a clear understanding of the consumption limits for their specific exposure group. We have presented in the present study the maximum allowable fish intake (CR_{max}) based on the measured Hg level for each studied species and calculated the number of meals per month basis (CR_{mm}) to better educate fish consumers.

The highest number of recommended monthly meals was observed for *T. albacares*, with up to 11 and 5 meals per month for adults and children, respectively. We consider this species the best choice of seafood among the studies species in the present study. In fact, *T. albacares* is one of the most consumed tuna species in the world, with a global production of around 1.57 million tons produced in 2020, occupying 7th place overall among fishery captures (FAO, 2022).

For other species, adults (70 kg) can eat up to 8 and 6 meals per month, while children (15 kg) can eat up to 4 and 2 meals per month of *C. hippurus* and *I. albicans*, respectively. Adults should eat up to 4, 3 and 2 meals per month, while children should eat up to 2, 1 and 1 meals per month of *T. obesus*, *T. alalunga* and *P. glauca*, respectively. Finally, adults should eat up to 1 meal per month, while children should avoid consumption of *I. oxyrinchus* and *X. gladius*. This might be problematic for regions with traditional and widespread seafood consumption.

Another potential issue is the mislabeling of seafood. In Brazil, for example, many shark species can be commercialized under the label of "cação", which limit the consumer knowledge of the actual species being consumed (Bernardo et al., 2020). Brazil is the 11th largest producer of sharks, the 17th largest exporter of shark fins, and the single largest importer of shark meat globally (Barreto et al., 2017; FAO, 2020). The volume of *P. glauca* imported by Brazilian industries across the country is comparable to the total domestic production of sharks and rays combined, amounting to approximately 21,000 t (Barreto et al., 2017). Therefore, shark meat consumption might be exposing human population in Brazil, as recently suggested by Campos et al. (2024), and worldwide, to excessive Hg levels with the potential of causing harmful health effects.

5. Conclusions

The present study shows Hg concentrations in several species of tuna, tuna-like and sharks caught in the Western Equatorial Atlantic Ocean. We identified differences among species, with significantly higher Hg concentrations in sharks (*I. oxyrinchus* and *P. glauca*) and Swordfish (*X. gladius*) compared to the Atlantic sailfish (*I. albicans*), the Albacore (*T. alalunga*) and the Bigeye (*T. obesus*). The species Yellowfin tuna (*T. albacares*) and Dolphinfish (*C. hipurrus*) presented the lowest concentrations among the studied species. Of the fish that were analyzed for MeHg, all showed high percentages compared to THg, corroborating the global scenario for these species. Furthermore, our results suggest that oceanic pelagic fish from the South Atlantic tend to have higher mercury concentrations by fish weight compared to those from the North Atlantic.

An extensive stable isotope analysis (SIA) was carried out in which significant differences were found between the species, which showed the same trend for Hg, i.e. the highest $\delta^{15}N$ and $\delta^{13}C$ values were observed for sharks and Swordfish. These results show that although the eight species studied theoretically occupy the same trophic levels, there are relevant differences associated with distinct habitats, diets and foraging areas. These conditions certainly have an influence on the accumulation of contaminants, such as Hg, and partly explain the variability in accumulation between species.

Finally, it is crucial to highlight that consuming fish offers significant health benefits for humans, as fish are an integral part of a balanced diet and are rich in Omega-3 fatty acids. However, the recommendations provided in this study are important for ensuring the safe consumption of this species by humans and may indicate a significant exposure to Hg. One example of the importance of this study was that children should avoid consumption of shark meat if possible. We recognize that fish consumption is one of the healthiest food choices and that assessing the risks and benefits mainly depends on which species are being selected and how often they are consumed.

CRediT authorship contribution statement

Felipe A. de Alencar Goyanna: Writing – original draft, Methodology, Investigation, Formal analysis, Data curation, Conceptualization. **Moisés Fernandes Bezerra:** Writing – review & editing, Supervision, Investigation, Formal analysis. **Guelson Batista da Silva:** Writing – review & editing, Resources, Conceptualization. **Carlos Eduardo de Rezende:** Resources, Methodology. **Wanderley Rodrigues Bastos:** Resources, Methodology. **Luiz Drude de Lacerda:** Writing – review & editing, Supervision, Resources, Funding acquisition, Conceptualization.

Declaration of competing interest

The authors declare the following financial interests/personal relationships which may be considered as potential competing interests: Bezerra, M.F. reports financial support was provided by Fundação Cearense de Apoio ao Desenvolvimento Científico e Tecnológico (FUNCAP). Lacerda, L.D. reports financial support was provided by Conselho Nacional de Desenvolvimento Científico e Tecnológico - CNPq. Rezende, C.E. reports financial support was provided by Fundação de Amparo à Pesquisa do Estado do Rio de Janeiro - FAPERJ. If there are other authors, they declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.scitotenv.2025.178994.

Data availability

Data will be made available on request.

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