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FELIPE AUGUSTO DE ALENCAR GOYANNA

MERCÚRIO (Hg), ISÓTOPOS ESTÁVEIS (δ^{15}N & δ^{13}C) E RECOMENDAÇÕES DE SEGURANÇA ALIMENTAR PARA TUBARÕES E PEIXES PELÁGICOS DE NÍVEL TRÓFICO SUPERIOR NO OCEANO ATLÂNTICO

FORTALEZA
2024

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Orientador: Prof. Dr. Luiz Drude de Lacerda.
Coorientador: Prof. Dr. Moisés Fernandes Bezerra.

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A Deus e minha amada família.

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“O pescador tem dois amor
Um bem na terra
Um bem no mar...” (DORIVAL CAYMMI, 1978).

RESUMO

Peixes oceânicos predadores de alto nível trófico são utilizados como espécies sentinelas na avaliação da presença de contaminantes persistentes nos oceanos de todo o mundo, como por exemplo o mercúrio (Hg). Além disso, são espécies que são consumidas globalmente por humanos, que eventualmente podem ser expostos à intoxicação alimentar. Nesta perspectiva, a tese tem como objetivo realizar uma análise das concentrações de Hg e as proporções de nitrogênio ($^{15}N/^{14}N$) e carbono ($^{13}C/^{12}C$) em relação a um padrão de referência em espécies de peixes oceânicos capturados no oceano Atlântico Equatorial. O primeiro capítulo foi destinado a realizar uma ampla revisão em todo o Atlântico e Mediterrâneo, na qual foram selecionadas diversas publicações, entre 1973 e 2022, analisadas e devidamente compiladas, com o objetivo de investigar a contaminação por Hg em nove espécies de peixes ósseos, *Thunnus thynnus*, *Thunnus albacares*, *Thunnus obesus*, *Thunnus atlanticus*, *Thunnus alalunga*, *Katsuwonus pelamis*, *Xiphias gladius*, *Coryphaena hippurus* e *Euthynnus alletteratus*, bem como duas espécies de tubarões, *Prionace glauca* e *Isurus oxyrinchus*. Foi verificada diferença significativa entre tubarões e peixes ósseos, com concentrações mais altas de Hg nos tubarões. As regiões de ocorrência foram divididas em quatro grandes áreas, mas não foram observadas diferenças significativas entre regiões. Das espécies selecionadas, nove apresentaram indivíduos com valores de concentração de Hg que excederam os limites de segurança estabelecidos pelas agências de saúde em todo o mundo. O segundo capítulo foi destinado a reportar o primeiro registro de concentrações de Hg e isótopos estáveis ($\delta^{13}C$ e $\delta^{15}N$) em *T. alalunga* capturadas no Oceano Atlântico Oeste Equatorial, tendo em vista que se trata de uma das principais espécies de atum consumidas no mundo. Foi verificado que as concentrações de Hg em *T. alalunga*, sendo 92% de metil-Hg, são mais altos do que em outras sub-regiões do Oceano Atlântico, apesar de seu tamanho corporal menor. As concentrações de Hg encontradas são semelhantes às dos oceanos Pacífico e Índico, mas inferiores às do Mediterrâneo. Esses resultados são discutidos considerando as possíveis diferenças nos valores de isótopos estáveis ($\delta^{15}N$ e $\delta^{13}C$) das populações de *T. alalunga* de várias áreas oceânicas e em comparação com outras espécies de atum em todo o mundo. O terceiro capítulo é destinado a reportar as análises de Hg e isótopos estáveis ($\delta^{15}N$ e $\delta^{13}C$), bem como apresentar recomendações de consumo em oito espécies de peixes oceânicos predadores capturados no Oceano Atlântico Oeste Equatorial. Foram encontradas diferenças significativas nas concentrações de Hg e isótopos entre as espécies. As concentrações de Hg foram mais altas em *I. oxyrinchus*, *X. gladius* e *P. glauca*, respectivamente, intermediárias em *T. alalunga*, *Istiophorus albicans* e *T. obesus,* enquanto que as concentrações de Hg foram mais baixas em *T. albacares* e *C. hippurus*. Os valores mais altos de $\delta^{15}N$ foram observados em *I. oxyrinchus*, *X. gladius* e *P. glauca*, em comparação com *C. hippurus*, *I. albicans* e *T. albacares*, que apresentaram os valores mais baixos. Valores intermediários foram observados em *T. obesus* e *T. alalunga*. Os valores mais altos de $\delta^{13}C$ foram observados em *C. hippurus* em comparação com *T. alalunga*, *P. glauca* e *X. gladius*. Somente *I. oxyrinchus, P. glauca* e *X. gladius* ultrapassaram o limite máximo regional de 1.000 $ng.g^{-1}$ de Hg, estabelecido na legislação brasileira. Finalmente, foram realizadas recomendações de consumo mensal por humanos para cada espécie estudada. Os valores médios de consumo seguro variaram de 22,3 ± 23,6 $g.dia^{-1}$ para adultos a 4,8 ± 5,0 g dia-1 para crianças em consumidores gerais. A taxa de consumo foi maior em *T. albacares* e *C. hippurus*, menor em *I. oxyrinchus* e *X. gladius*. O número estimado de refeições por mês variou entre 0 a 11, considerando uma porção de 150 e 75 g para adultos e crianças, respectivamente. As crianças devem evitar o consumo das espécies *X. gladius* e *I. oxyrinchus*.

Palavras-chave: mercúrio; isótopos; tubarões; atuns; contaminação; Atlântico.

ABSTRACT

High trophic level predatory ocean fish are used as sentinel species to assess the presence of persistent contaminants in the world's oceans, such as mercury (Hg). In addition, they are species that are consumed globally by humans, who may eventually be exposed to food poisoning. With this in mind, the thesis aims to analyze Hg concentrations and the proportions of nitrogen ($^{15}N/^{14}N$) and carbon ($^{13}C/^{12}C$) in relation to a reference standard in oceanic fish species caught in the Equatorial Atlantic Ocean. The first chapter was designed to carry out a broad review across the Atlantic and Mediterranean, in which several publications were selected between 1973 and 2022 were selected, analyzed and duly compiled, with the aim of investigating Hg contamination in nine species of bony fish, *Thunnus thynnus, Thunnus albacares, Thunnus obesus, Thunnus atlanticus, Thunnus alalunga, Katsuwonus pelamis, Xiphias gladius, Coryphaena hippurus* and *Euthynnus alletteratus*, as well as two shark species, *Prionace glauca* and *Isurus oxyrinchus*. A significant difference was found between sharks and bony fish, with higher Hg concentrations in sharks. The regions of occurrence were divided into four large areas, but no significant differences were observed between regions. Of the species selected, nine had individuals with Hg concentration values that exceeded the safety limits set by health agencies worldwide. The second chapter was designed to report the first record of Hg concentrations and stable isotopes ($\delta^{13}C$ and $\delta^{15}N$) in *T. alalunga* caught in the Western Equatorial Atlantic Ocean, given that it is one of the main tuna species consumed worldwide. It was found that Hg concentrations in *T. alalunga*, 92% of which are Methyl-Hg, are higher than in other sub-regions of the Atlantic Ocean, despite its smaller body size. The Hg concentrations found are similar to those in the Pacific and Indian Oceans, but lower than those in the Mediterranean. These results are discussed considering the possible differences in stable isotope values ($\delta^{15}N$ and $\delta^{13}C$) of *T. alalunga* populations from various oceanic areas and in comparison with other tuna species around the world. The third chapter is designed to report Hg and stable isotope ($\delta^{15}N$ and $\delta^{13}C$) analyses, as well as present consumption recommendations in eight species of predatory oceanic fish caught in the Western Equatorial Atlantic Ocean. Significant differences in Hg and isotope concentrations were found between species. Hg concentrations were highest in *I. oxyrinchus*, *X. gladius* and *P. glauca*, respectively, intermediate in *T. alalunga*, *Istiophorus albicans* and *T. obesus*, while Hg concentrations were lowest in *T. albacares* and *C. hippurus*. The highest $\delta^{15}N$ values were observed in *I. oxyrinchus*, *X. gladius* and *P. glauca*, compared to *C. hippurus*, *I. albicans* and *T. albacares*, which showed the lowest values. Intermediate values were observed in *T. obesus* and *T. alalunga*. The highest $\delta^{13}C$ values were observed in *C. hippurus* compared to *T. alalunga, P. glauca* and *X. gladius*. Only *I. oxyrinchus, P. glauca* and *X. gladius* exceeded the regional maximum limit of 1,000 $ng.g^{-1}$ of Hg established by Brazilian legislation. Finally, recommendations for monthly consumption by humans were made for each species studied. The average safe consumption values ranged from 22.3 ± 23.6 $g.day^{-1}$ for adults to 4.8 ± 5.0 g day^{-1} for children in general consumers. The consumption rate was higher in *T. albacares* and *C. hippurus*, lower in *I. oxyrinchus* and *X. gladius*. The estimated number of meals per month ranged from 0 to 11, considering a portion of 150 and 75 g for adults and children, respectively. Children should avoid eating *X. gladius* and *I. oxyrinchus*.

Keywords: mercury; isotopes; sharks; tuna; contamination; Atlantic.

LISTA DE FIGURAS

CAPÍTULO 1

CAPÍTULO 2

CAPÍTULO 3

LISTA DE TABELAS

CAPÍTULO 2

CAPÍTULO 3

SUMÁRIO

INTRODUÇÃO

Um dos principais problemas gerados pelo antropoceno é a contaminação dos oceanos a nível global, impactando diretamente sobre saúde dos ecossistemas marinhos (ALI *et al*., 2019). Neste contexto, a preocupação com metais não é algo tão recente no mundo, muitos trabalhos e pesquisas científicas foram e estão sendo desenvolvidos desde o início do século passado abordando a assunto. Dentro dessa classe de contaminantes, destaca-se o mercúrio (Hg) como sendo um dos mais tóxicos para os organismos vivos em geral, inclusive para os humanos, que estão sujeitos a sua bioacumulação durante seu ciclo de vida, que por sua vez contribuem globalmente com as emissões derivadas da atividade antrópica, alterando o ciclo biogeoquímico do metal (DRISCOLL et al., 2013; GWOREK *et al*., 2016; STERN *et al*., 2012; UNEP, 2018).

Nos oceanos, o tempo de resiliência do Hg pode variar de trinta anos, em profundidades menores ($\geq$ 200m) até mais um século em regiões mais profundas (UNEP, 2018), isso mostra que embora a maior parte esteja em profundidades menores o percentual em profundidades maiores é relevante. Em áreas oceânicas pelágicas, a principal fonte de Hg é a deposição atmosférica, predominantemente na forma de Hg^{2+} que sofre metilação biótica e abiótica, na zona fótica, formando metilmercúrio (CH_3Hg) (OBRIST *et al*., 2018). O metilmercúrio presente no ecossistema marinho pode ser assimilado na cadeia alimentar por meio da absorção direta da água no fitoplâncton, de onde é transferido para os organismos consumidores por meio da alimentação (GOSNELL *et al*., 2021). As assimilações menos eficientes incluem a absorção pelas brânquias dos peixes e a deposição durante a síntese das estruturas duras dos invertebrados (SAIDON *et al*., 2024).

O Hg sofre biomagnificação nas teias alimentares aquáticas, ou seja, aumenta sua concentração à medida que eleva o nível trófico dos organismos, isso resulta em predadores de nível superior com concentrações bem mais altas do que os produtores primários. Tais concentrações são deletérias aos organismos e mesmo que estejam expostos a baixas concentrações de Hg no ambiente (0,08 – 6,0 $\mu g.g^{-1}$), trabalhos com assimilação de Hg pela alimentação em aves, peixes, mamíferos e répteis evidenciam uma série de disfunções nos sistemas endócrino, reprodutivo e imunológico dos animais (HAMMERSCHMIDT *et al*., 2002; SCHNEIDER *et al*., 2013; SPALDING *et al*., 2000).

Em geral, peixes pelágicos de nível superior podem refletir variações espaciais na contaminação por Hg em várias regiões oceânicas, o que pode ajudar a entender o destino ambiental do Hg no ecossistema marinho (GOYANNA *et al*., 2023) e destacar a importância de avaliar várias espécies em esforços de biomonitoramento (MÉDIEU *et al*., 2023). O monitoramento de longo prazo nesses peixes também é limitado e as informações atuais sobre o acúmulo de Hg mostram níveis maiores do que os esperados, considerando as mudanças nas emissões atmosféricas globais e na deposição (MÉDIEU *et al.*, 2023).

A distribuição de mercúrio (Hg) em escala global nos grandes peixes carnívoros oceânicos, principalmente atuns, espécies semelhantes a atuns e tubarões, mostra que as variações nos níveis de mercúrio são parcialmente impulsionadas por características ecológicas específicas das espécies, como profundidade de forrageamento, composição da dieta e tamanho do corpo (LACERDA *et al.*, 2017; SCHNEIDER *et al.*, 2013), o que sugere que grandes peixes ósseos e tubarões podem fornecer informações complementares sobre a distribuição vertical e horizontal do metal no oceano (GOYANNA et al, 2023; MÉDIEU *et al.*, 2023). Portanto, para usar esses peixes como biomonitores dos níveis de Hg, de acordo com as Convenções de Minamata sobre Mercúrio, é necessário entender os fatores ecológicos da acumulação de Hg e separá-los da variabilidade espacial nas concentrações de Hg, tanto verticalmente na coluna d'água quanto horizontalmente entre as regiões oceânicas. Além disso, ainda há lacunas significativas nos dados de acúmulo de Hg para muitas espécies pelágicas de regiões oceânicas no Oceano Atlântico Sul (GOYANNA *et al.*, 2023; MÉDIEU *et al.*, 2023).

Os fatores ecológicos frequentemente descritos para influenciar o acúmulo de Hg na megafauna marinha são o habitat de alimentação das espécies, incluindo a profundidade; a ecologia da alimentação e as mudanças ontogenéticas associadas; as interações tróficas (predação) e a estrutura da teia alimentar (BARRIOS-RODRIGUEZ *et al.*, 2024; MOURA *et al.*, 2020). Todas essas características podem ser avaliadas por meio da Análise de Isótopos Estáveis (SIA) de elementos leves, como carbono e nitrogênio, em tecidos de organismos marinhos (PETERSON; FRY, 1987). As proporções de nitrogênio (^{15}N /^{14}N) e carbono (^{13}C / ^{12}C) em relação a um padrão de referência (doravante $\delta^{15}N$ e $\delta^{13}C$, respectivamente) são mais comumente usadas para examinar vários processos tróficos (POST *et al.*, 2007; YEAKEL *et al.*, 2016). Por exemplo, como o $\delta^{13}C$ muda muito pouco à medida que o carbono se move da base da teia alimentar para os consumidores de nível superior (enriquecimento de ~0,5‰-1‰) e reflete as principais fontes de carbono para a teia alimentar em questão (BOUILLON *et al.*, 2011; PETERSON; FRY, 1987). Por outro lado, o $\delta^{15}N$ é enriquecido nos consumidores (~2‰ - 4‰) em relação à sua dieta, o que pode fornecer uma estimativa da posição trófica e caracterização do nicho trófico (HUSSEY *et al.*, 2010; LAYMAN *et al.*, 2007; POST, 2002).

Portanto, o uso da SIA pode complementar a avaliação do acúmulo de Hg em peixes com níveis tróficos mais altos. Como o $\delta^{15}N$ geralmente aumenta com níveis tróficos mais altos e o Hg também se acumula nesses níveis, pode-se observar uma correlação positiva entre os valores de $\delta^{15}N$ e a concentração de Hg nos organismos. Isso é especialmente útil para quantificar o fator de biomagnificação trófica (TMF) do mercúrio na teia trófica, fornecendo informações sobre a magnitude dos aumentos de Hg em vários níveis tróficos (CONDINI *et al.*, 2017; SINKUS *et al.*, 2017).

A produção média global de organismos é de cerca de 178 milhões de toneladas (t) por ano, sendo que a captura de peixes marinhos responde por aproximadamente 46% (81,1 milhões de toneladas) desse montante, o que é maior do que a aquicultura (terrestre e marinha) e as capturas continentais (FAO, 2022). Estimativas sugerem que 88,4% da produção média é destinada ao consumo humano, resultando em um consumo aparente per capita de 20,4 kg por ano (ou 55,9 gramas por dia) (FAO, 2022). As capturas globais de atum e espécies semelhantes ao atum atingiram 7,8 milhões de toneladas em 2020, sendo que o Bonito listrado (*Katsuwonus pelamis*) e Albacora laje (*Thunnus albacares*) responderam por 55% das capturas desse grupo (FAO, 2022). Apesar de ter um sistema de gestão de pesca altamente deficitário (NETO *et al.*, 2021), o Brasil é um ator importante na atividade pesqueira realizada no Oceano Atlântico, com capturas totais que atingiram 52.519 t em 2021, incluindo atum e outras espécies pelágicas semelhantes (ICCAT, 2022). Especificamente, as capturas brasileiras totais em 2021 por tipo de espécie foram de 4.629 t para o Tubarão-azul, *Prionace glauca*; 2.110 t para o Espadarte, *Xiphias gladius*; 6.499 t para o Albacora bandolim, *Thunnus obesus*; 13.664 t para Albacora laje, *T. albacares*; 516 t para a Albacora branca, *Thunnus alalunga*; 477 t para o Tubarão mako, *Isurus Oxyrinchus* e 24 t para o Agulhão vela do Atlântico, *Istiophorus albicans* (ICCAT, 2022).

Em relação ao risco de exposição humana ao Hg, o consumo de peixe é a principal via de exposição crônica ao Hg. As avaliações de risco geralmente mostram que fatores como tipo e tamanho da espécie, frequência de consumo e níveis de Hg são os fatores mais importantes que regem a exposição humana por meio do consumo de peixes (BEZERRA *et al.*, 2023), mas as avaliações de exposição ainda são limitadas para essas grandes espécies de peixes pelágicos (GOYANNA *et al.*, 2023). Além disso, os níveis de Hg nessas espécies geralmente estão na forma orgânica e mais tóxica de metilmercúrio (~95%) (MONTEIRO; LOPES, 1990; STORELLI; MARCOTRIGIANO, 2001). Estima-se que os peixes de nível trófico superior da costa atlântica equatorial brasileira, incluindo a área de descarga do rio Amazonas, estejam entre os mais altos em concentração de metilmercúrio em todo o mundo (WU; ZHANG, 2023). Como resultado, apesar de o Brasil responder por menos de 1% das capturas globais de peixes (FAO, 2022), sua contribuição estimada para as exportações de metilmercúrio por meio da captura de peixes representa 21% do total global. (WU; ZHANG, 2023).

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HIPÓTESES CIENTÍFICAS

A hipótese principal é que não somente o peso, a idade e o comprimento corporal das espécies predadoras de alto nível trófico são responsáveis pelas concentrações de Hg nos seus tecidos musculares, mas que as variações ecológicas e ambientais, também influenciam na concentração de Hg dessas espécies. Essa hipótese foi testada considerando os dados globais de Hg relatados anteriormente sobre atum e espécies semelhantes. Foi levantada a hipótese de que os níveis de Hg em peixes pelágicos de nível trófico superior do Atlântico Oeste Equatorial diferem entre as espécies e são menores em comparação com espécies semelhantes que ocorrem em outras regiões oceânicas.

Considerando que isótopos estáveis de carbono e nitrogênio fornecem informações úteis sobre o habitat de alimentação, a dieta e o nível trófico, levantamos a hipótese de que o nicho isotópico e outras características ecológicas refletidas pelo carbono 13 e nitrogênio 15 explicam parcialmente a variabilidade no acúmulo de Hg entre as espécies.

Finalmente, considerando a alta posição trófica desses peixes e a conhecida biomagnificação do Hg nas redes alimentares marinhas, nossa hipótese é que os níveis de Hg nesses grandes peixes pelágicos estão em uma faixa potencialmente prejudicial aos consumidores humanos.

OBJETIVOS

Objetivo Geral

Ampliar a compreensão da distribuição de Hg nas principais espécies de peixes oceânicos de interesse comercial na região do Oceano Atlântico Oeste Equatorial (*Coryphaena hippurus, Euthynnus alletteratus, Istiophorus albicans, Isurus oxyrinchus, Prionace glauca, Thunnus albacares, Thunnus alalunga, Thunnus atlanticus, Thunnus obesus, Thunnus thynnus, Xiphias gladius* e *Katsuwonus pelamis)* e seus fatores (biológicos, ecológicos e ambientais) controladores.

Objetivos específicos

- Realização de ampla revisão dos principais trabalhos determinando as concentrações de Hg em espécies de grandes peixes pelágicos oceânicos, capturadas no Oceano Atlântico, comparando a distribuição entre espécies, bem como entre regiões, possíveis causas e efeitos tóxicos

- Quantificar e avaliar o acúmulo de Hg em peixes pelágicos de nível trófico superior no Oceano Atlântico Oeste Equatorial, incluindo a realização de correlações, como a relação com o tamanho do corpo e a variabilidade associada às espécies.

- Avaliar os fatores ecológicos do acúmulo de Hg usando a análise de isótopos estáveis de carbono e nitrogênio.

- Estimar a exposição ao Hg por meio do consumo de peixes, fazendo recomendações favoráveis ao consumidor sobre as melhores opções de consumo em nossa área de estudo.

CAPÍTULO I: MERCÚRIO EM TUBARÕES E PEIXES ÓSSEOS OCEÂNICOS DE NÍVEL TRÓFICO SUPERIOR - UMA REVISÃO SISTEMÁTICA[1]

Resumo: As atividades antropogênicas contribuem para quase metade das emissões atuais de Hg na atmosfera. No habitat marinho, os peixes predadores oceânicos bioacumulam Hg ao longo de suas vidas, tornando seu consumo a principal via de exposição ao Hg em seres humanos. Nesse contexto, várias publicações, entre 1973 e 2022, foram selecionadas, analisadas e devidamente compiladas, com o objetivo de investigar a contaminação por Hg em nove espécies de peixes ósseos: *Thunnus thynnus* (8 publicações), *Thunnus albacares* (19), *Thunnus obesus* (7), *Thunnus atlanticus* (5), *Thunnus alalunga* (4), *Katsuwonus pelamis* (8), *Xiphias gladius* (18), *Coryphaena hippurus* (7) e *Euthynnus alletteratus* (4), bem como duas espécies de peixes cartilaginosos *Prionace glauca* (13 publicações) e *Isurus oxyrinchus* (8). Esses estudos totalizaram 5973 indivíduos. Classificamos as espécies de acordo com os grupos taxonômicos e a região de captura e encontramos uma diferença significativa entre tubarões e peixes ósseos, com concentrações mais altas de Hg nos tubarões. As regiões de ocorrência foram divididas em quatro grandes áreas (Atlântico Norte - NAO, Atlântico Sul - SAO, Oceano Atlântico Equatorial - EAO e Mediterrâneo - MED), mas não foram observadas diferenças significativas ao comparar as concentrações gerais de Hg em peixes entre as regiões (incluindo todas as espécies). . Além disso, foi realizada uma discussão aprofundada dos riscos associados ao consumo humano dessas espécies, já que nove das espécies selecionadas apresentaram indivíduos com valores de concentração de Hg que excederam os limites de segurança (1 ppm) estabelecidos pelas agências de saúde em todo o mundo.

Palavras-Chave: Mercúrio, Tubarões, Atlântico, Atum, Poluição, Contaminação.

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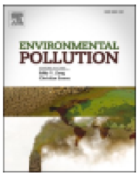

Review

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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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.

Keywords: Mercury, Sharks, Atlantic, Tuna, Pollution, Contamination

Introdution

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 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.

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).

Statistical Analysis

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).

Results and Discussion

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).

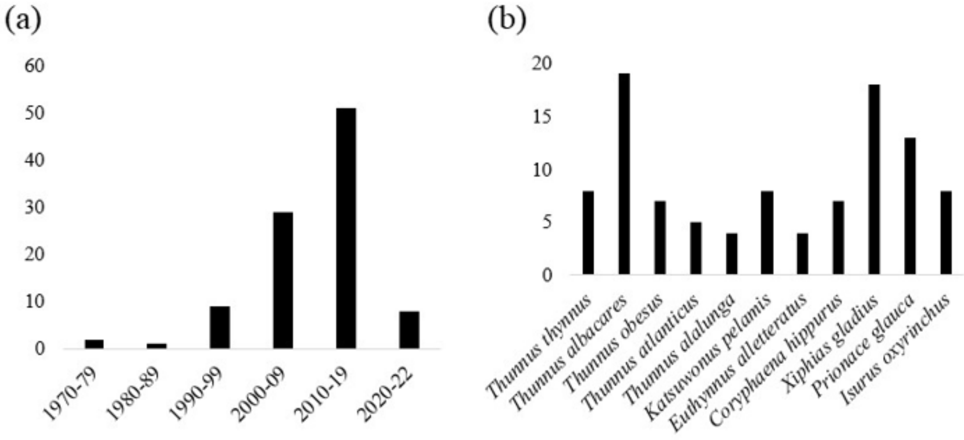

Figure 1 - Distribution of publications in different decades (a) and the number of publications by species (b).

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.

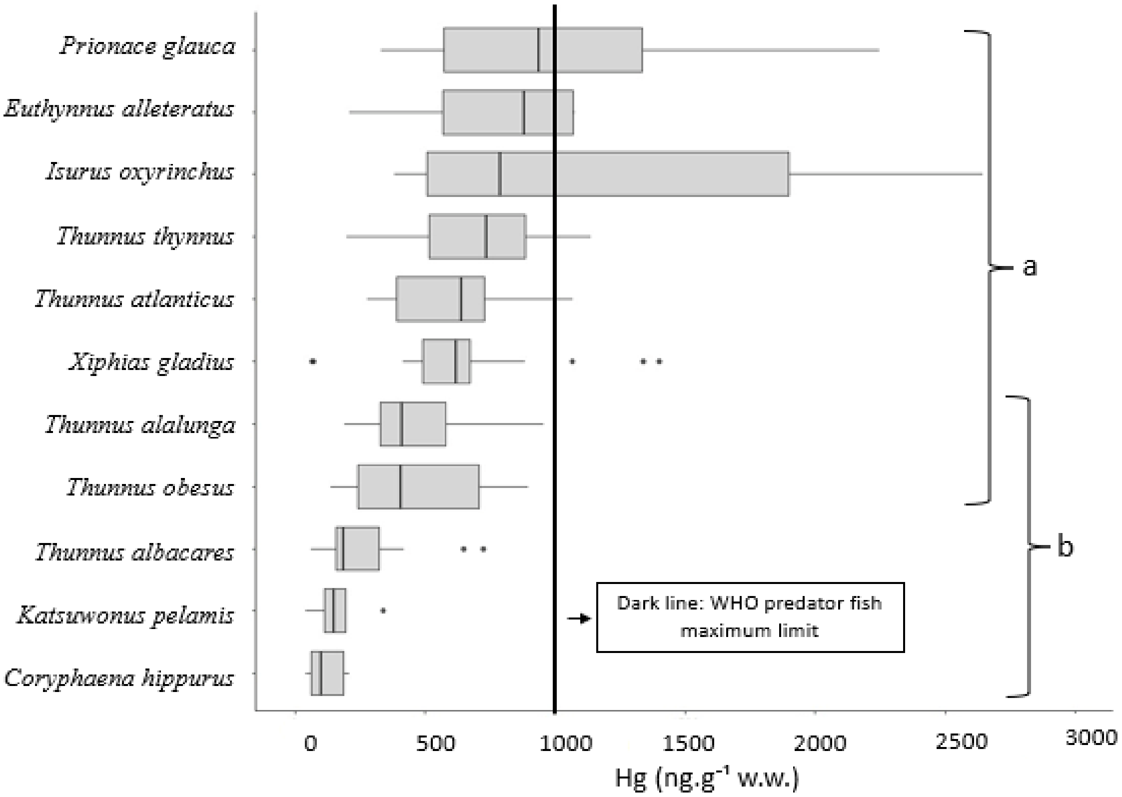

Figure 2 - Boxplot with Hg variations in 11 species of oceanic fish from Atlantic and Mediterranean. Letters a and b represent statistically significant differences

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 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.

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). Figure 3 presents a boxplot of the Hg variation among the four oceanic regions studied for the respective species.

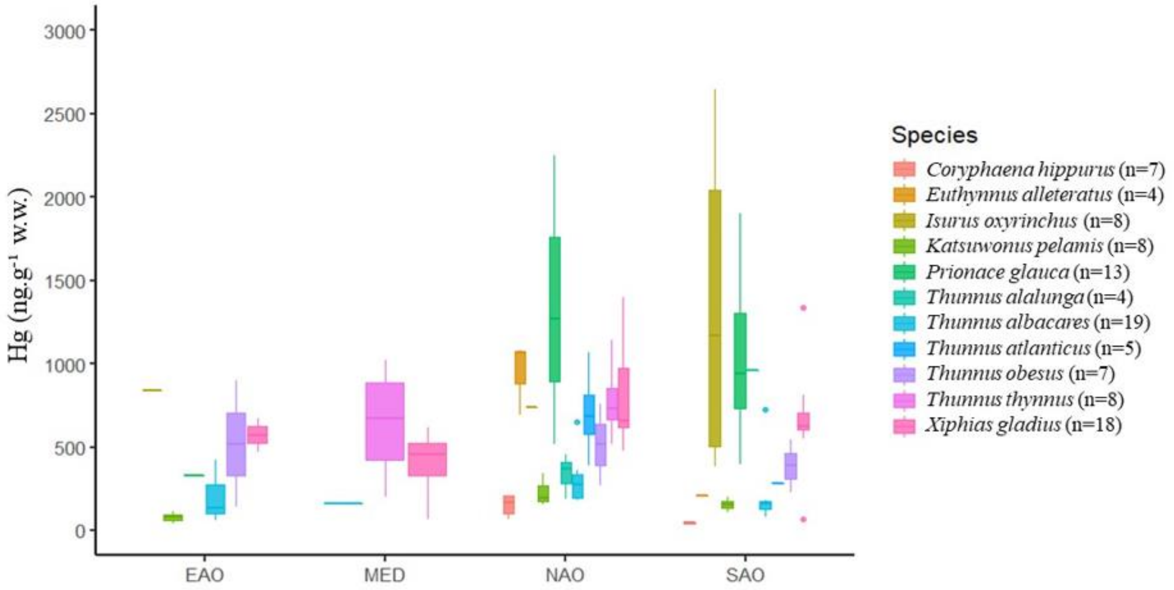

Figure 3. Boxplot of variation of Hg concentrations in oceanic fish species in the respective capture areas. EAO – Equatorial Atlantic Ocean; MED – Mediterranean; NAO – North Atlantic Ocean; SAO – South Atlantic Ocean.

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, 2013). 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.

Taxonomical differences in Hg concentrations

According to the reviewed papers, we divided the studied species into different Class, 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) (Figure 4).

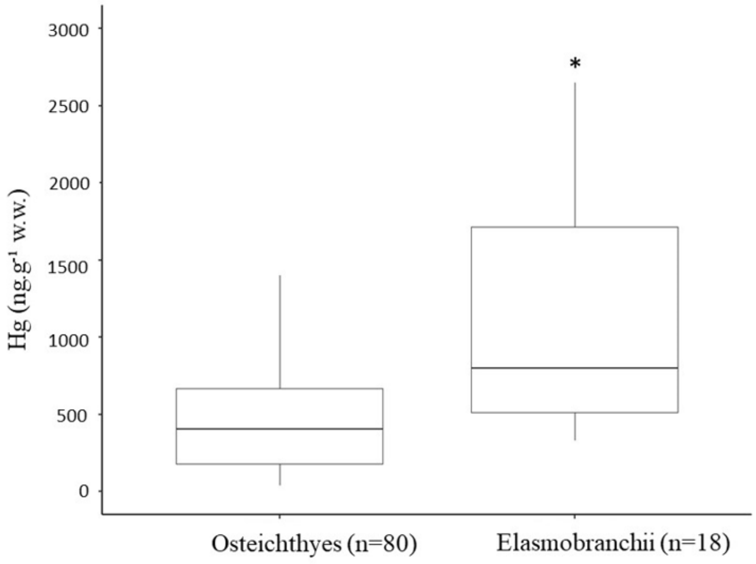

Figure 4 - Boxplot of variation of Hg concentrations in Osteichthyes and Elasmobranchii considering all catch areas. *Statistically significant differences.

When considering the overall Hg averages (considering all geographic areas), we found a significant higher concentration in Elasmobranchii compared to Osteichthyes (Figure 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). Highest Hg concentrations in elasmobranchs compared to bony fish, was 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; Pierce and Bennett, 2010, Tiktak et al, 2020, Bradley et al., 2017; Trudel and Rasmussen, 1997; Dang and Wang, 2012). 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.

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). Global catch includes about 1.700 exploited species, of which finfish account for about 85% of total marine catch production, with small pelagics being the main group, followed by gadiforms and tuna 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 1,000 $ng.g^{-1}$ (FAO/WHO, 1972). 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. alltteratus, I. oxyrinchus, T. thynnus, T. atlanticus and X. gladius*) exceeded the legal limits, thus making some fish unfit for human consumption (Figure 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 1.000 $ng.g^{-1}$. In the Mediterranean, for example, 44% of 169 individuals of *T. thynnus* caught presented Hg concentrations higher than 1,000 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 average consumption of freshwater fish (capture and aquaculture) is quite low at 3.95 kg per capita per year in 2013 (FAO, 2017), 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, 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 1.000 $ng.g^{-1}$ could possibly be harmful.

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.

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CAPÍTULO II: PRIMEIRO REGISTRO DE CONCENTRAÇÕES DE MERCÚRIO E ISÓTOPOS ESTÁVEIS (^{13}C & ^{15}N) EM ALBACORA BRANCA (THUNNUS ALALUNGA) DO OCEANO ATLÂNTICO EQUATORIAL OCIDENTAL[2]

Resumo: Este artigo relata o primeiro registro de concentrações de mercúrio total (THg) em albacora branca (*Thunnus alalunga*), uma das principais espécies de atum capturadas no Oceano Atlântico Equatorial Atlântico Equatorial Ocidental e apresenta uma comparação preliminar com outras regiões e espécies de atum e espécies de atum. Média, desvio padrão e intervalo de concentrações em T. alalunga (515 ± 145 ng g^{-1} ww; 294 - 930 ng g^{-1} ww), sendo 92% de metil-Hg, são maiores do que em albacora de outras sub-regiões do Oceano Atlântico, apesar de seu tamanho corporal menor. Essas concentrações são semelhantes as verificadas nos oceanos Pacífico e Índico, mas inferiores que as observadas do Mediterrâneo. Em comparação com outras espécies simpátricas de atum, as concentrações são mais altas do que as do *T. albacares* e semelhantes às do *T. obesus*. Esses resultados são discutidos considerando as possíveis diferenças nos valores de isótopos estáveis (^{13}C e ^{15}N) das populações de *T. alalunga* de várias áreas oceânicas e comparadas a outras espécies de atum em todo o mundo.

Palavras-Chave: Mercúrio, Albacora, Atlântico, Atum, Poluição, Contaminação.

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Baseline

First record of mercury concentrations and stable isotopes (^{13}C & ^{15}N) in albacore (*Thunnus alalunga*) from the Western Equatorial Atlantic Ocean

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ABSTRACT

This paper reports the first record of total mercury (THg) concentrations in albacore (*Thunnus alalunga*), one of the main tuna species caught from the Western Equatorial Atlantic Ocean and presents a preliminary comparison with other regions and tuna species. Mean, standard deviation and range of concentrations in *T. alalunga* (515 ± 145 ng g^{-1} ww; 294–930 ng g^{-1} ww) with 92 % being of methyl-Hg, are higher than in albacore from other Atlantic Ocean subregions despite their smaller body size. These concentrations are similar to those from the Pacific and Indian oceans, but lower than in the Mediterranean. Compared to other sympatric tuna species, concentrations are higher than those in *T. albacares* and similar to *T. obesus*. These results are discussed considering the potential differences in stable isotope values (^{13}C and ^{15}N) of *T. alalunga* populations from multiple oceanic areas and compared to other tuna species worldwide.

Mercury, without any known biological function, is a potent neurotoxin and is globally distributed in the oceans that bio-accumulates and biomagnifies through the food web. The biomagnification process results in top level predators presenting Hg concentrations at levels potentially harmful to the organism and, ultimately, to humans consuming these types of seafood. Tuna and tuna-like species are top consumers in oceanic food-webs and are among the most captured species by fishery activities potentially posing risks of Hg exposure in humans largely consuming this type of seafood. As long-life marine animals, such oceanic tuna and tuna-like fish are particularly exposed to environmental Hg concentrations. Their wide distribution and lifespan compatible with Hg residence time in ocean surface waters, make them reliable biological monitors of the long-term, large-scale, changes in Hg concentrations in the global ocean (Houssard et al., 2019; Médieu et al., 2023; Goyanna et al., 2023).

Two recent comprehensive reviews on global scale mercury (Hg) distribution in large oceanic carnivorous fish, mostly tuna and tuna-like (Goyanna et al., 2023; Médieu et al., 2023), have shown that these fishes reflect spatial trends of Hg contamination in their area of occurrence. Despite this, within the same geographical region, Hg concentrations vary largely among species which can be a response of species-specific ecological and biological traits, including foraging depth, diet composition, and body size (Lacerda et al., 2017). Therefore, to use these fishes as a biomonitor of Hg levels, in agreement with the Minamata Conventions on Mercury (UN Environment, 2019), it is necessary to disentangle these trait-responses from the actual Hg variability, both vertically through the water column and horizontally between oceanic regions. In addition, as highlighted by both review studies, there is a significant gap on Hg data for some species and oceanic regions, even for one of the most economically exploited tuna species, such as the Albacore *Thunnus alalunga*.

Albacore occurs in temperate and tropical waters of all oceans and the Mediterranean Sea (Collette and Nauen, 1983). It reaches sexual maturation at an age of approximately five years, corresponding to about 90 cm in length. This species migrates widely for feeding and breeding preferring temperatures between 10 °C and 20 °C, which, together with dissolved oxygen, influences their horizontal and vertical distributions. In the Atlantic Ocean, for example, vertical migrations reach 600 m (Graham and Dickson, 1981; Collette and Nauen, 1983). There are three distinct populations of Albacore in the Atlantic Ocean, one in the north, another in the south (separated by latitude 5°N), and one endemic of the Mediterranean Sea (ICCAT, 2006). Albacore is one of

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First record of mercury concentrations and stable isotopes (^{13}C & ^{15}N) in albacore (Thunnus alalunga) from the Western Equatorial Atlantic Ocean

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Abstract

This paper reports the first record of total mercury (THg) concentrations in albacore (*Thunnus alalunga*), one of the main tuna species caught from the Western Equatorial Atlantic Ocean and presents a preliminary comparison with other regions and tuna species. Mean, standard deviation and range of concentrations in *T. alalunga* (515 ±145 ng g^{-1} ww; 294 - 930 ng g^{-1} ww) with 92% being of methyl-Hg, are higher than in albacore from other Atlantic Ocean subregions despite their smaller body size. These concentrations are similar to those from the Pacific and Indian oceans, but lower than in the Mediterranean. Compared to other sympatric tuna species, concentrations are higher than those in *T. albacares* and similar to *T. obesus*. These results are discussed considering the potential differences in stable isotope values (^{13}C and ^{15}N) of *T. alalunga* populations from multiple oceanic areas and compared to other tuna species worldwide

Introdution

Mercury, without any known biological function, is a potent neurotoxin and is globally distributed in the oceans that bio-accumulates and biomagnifies through the food web. The biomagnification process results in top level predators presenting Hg concentrations at levels potentially harmful to the organism and, ultimately, to humans consuming these types of seafood.

Tuna and tuna-like species are top consumers in oceanic food-webs and are among the most captured species by fishery activities potentially posing risks of Hg exposure in humans largely consuming this type of seafood. As long-life marine animals, such oceanic tuna and tuna-like fish are particularly exposed to environmental Hg concentrations. Their wide distribution and lifespan compatible with Hg residence time in ocean surface waters, make them reliable biological monitors of the long-term, large-scale, changes in Hg concentrations in the global ocean (Houssard et al., 2019; Médieu et al., 2023; Goyanna et al., 2023).

Two recent comprehensive reviews on global scale mercury (Hg) distribution in large oceanic carnivorous fish, mostly tuna and tuna-like (Goyanna et al., 2023; Médieu etal., 2023), have shown that these fishes reflect spatial trends of Hg contamination in their area of occurrence. Despite this, within the same geographical region, Hg concentrations vary largely among species which can be a response of species-specific ecological and biological traits, including foraging depth, diet composition, and body size (Lacerda et al., 2017). Therefore, to use these fishes as a biomonitor of Hg levels, inagreement with the Minamata Conventions on Mercury (UN Environment, 2019), it is necessary to disentangle these trait-responses from the actual Hg variability, both vertically through the water column and horizontally between oceanic regions. In addition, as highlighted by both review studies, there is a significant gap on Hg data forsome species and oceanic regions, even for one of the most economically exploited tuna species, such as the Albacore *Thunnus alalunga*.

Albacore occurs in temperate and tropical waters of all oceans and the Mediterranean Sea (Collette and Nauen, 1983). It reaches sexual maturation at an age of approximately five years, corresponding to about 90 cm in length. This species migrates widely for feeding and breeding preferring temperatures between 10 °C and 20 °C, which, together with dissolved oxygen, influences their horizontal and vertical distributions. In the Atlantic Ocean, for example, vertical migrations reach 600 m (Graham and Dickson, 1981; Collette and Nauen, 1983). There are three distinct populations of Albacore in the Atlantic Ocean, one in the north, another in the south (separated by latitude 5°N), and one endemic of the Mediterranean Sea (ICCAT, 2006). Albacore is one of the four most exploited tuna species worldwide and is one of themain tuna species caught mainly because of its importance for the fish canning companies in the South Atlantic Ocean (Collette and Nauen, 1983).

Globally, albacore only accounts for about 5%, in weight, of the global tuna and tuna-like catches (FAO, 2018). In Brazil, the total catch of tuna and tuna-like fish in 2021 was52519 t, of which 561 t about 1.1% on a weight basis) were albacore but can be much larger depending on year as in 2002, when catches reached 7000 t (about 15% of the total) (ICCAT, 2022).

Wu and Zhang (2023), based on modelling, estimated that Brazil has the largest amount of methyl-Hg associated with fish catch (1285 kg yr^{-1}), accounting for 21% of the global total. According to these authors, most of this load comes from the high methyl-Hg content from Amazon fish (Castilhos et al., 1998; Bastos et al., 2015), but about 33% of the Brazilian Methyl-Hg catch come from large pelagic fish, primarily from the Central and Southwest Atlantic (Wu and Zhang, 2023). They linked this to the observed methyl-Hg concentrations in demersal fishes in the central Atlantic, near the mouth of the Amazon River and in top predators in the Brazilian Equatorial Atlantic Ocean which are among the highest globally.

Notwithstanding, the significant economic importance and the methyl-Hg contribution to the total load derived from Brazilian fisheries, Hg concentrations in T. alalunga from the Southwestern Atlantic has never been quantified before. In addition, in other regions of the Atlantic Ocean, this species has shown Hg concentrations higher than the acceptable limits for human consumption (Andersen & Depledge, 1997; Blasco et al., 2020), implicitly posing high threat of human exposure to Hg in other Atlantic Ocean subregions. In summary, most Hg data available for T. alalunga derive from the Pacific Ocean, with a few information from the North Atlantic and the Mediterranean Sea, but currently there is no data available from the Southwestern Atlantic (Médieu et al., 2023).

Stable isotope analysis (SIA) can be a valuable tool in Hg contamination assessments. The ratios of carbon (13C/12C, hereafter δ13C) and nitrogen (15N/14N, hereafter δ15N) are commonly used in ecological studies to understand trophic dynamics of wild populations (Peterson and Fry, 1987; Post et al., 2007). A stepwise increase in δ13C (~1‰) and δ15N (~2‰ - 4‰) values is observed in consumers compared to prey which reflect the different incorporation rate of lighter and heavier isotopes by the consumer’s metabolism during nutrient assimilation and tissue synthesis (Bouillon et al., 2011; Peterson and Fry, 1987). Considering the Hg is ubiquitous in marine ecosystems and diet is the most important route of uptake and accumulation in marine consumers (Hall et al., 1997), SIA can help understanding how Hg moves through marine food webs and accumulates in upper level predator (Al-Reasi et al., 2007; Moura et al., 2020).

This study reports, based on a relatively large sample size, the first record of Hg concentrations in T. alalunga from the Equatorial Western Atlantic Ocean. The observed results are compared with the Hg content found in T. alalunga from other oceanic regions and with other sympatric tuna species typically caught in the Atlantic Ocean.

Muscle samples of albacore were obtained directly in the industry during the landings from the commercial longline fleet of Natal, Rio Grande do Norte State in NE Brazil, during two seasons (August 2020; n = 4 and October 2022; n = 40). The vessels operate using pelagic longlines approximately 10 km long and 180 hooks attached (Viana et al., 2015). The samples were fished at geographical coordinates between 3° N - 6° S and 122 16°- 30° W (Figure1.). These catches are mostly constituted by *Thunnus albacares* (34%) and *Xiphias gladius* (31%), but *T. alalunga* represents about 4% of catches 124 totaling 2600 t in the past 20 years, but with over 2000 t in the past decade (Silva et al., 2016; Lira et al., 2017).

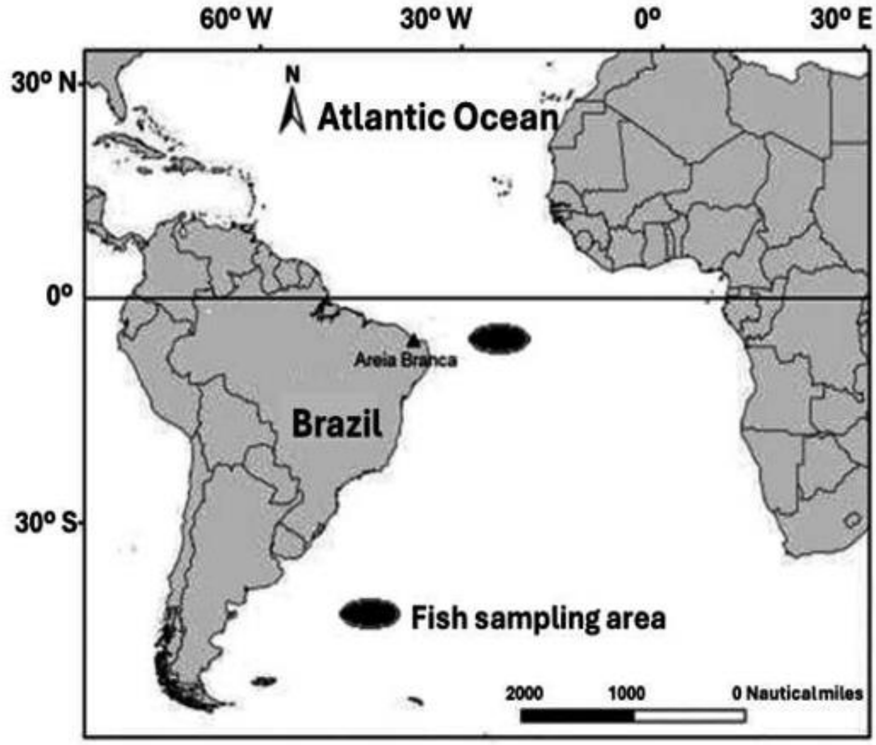


Figure 1. Map showing the approximate distribution of fishing areas from where T. alalunga was obtained for this study

All individuals were measured for total weight (kg) and samples of about 50 g of white muscle were taken from fresh whole fish, avoiding eviscerated individuals or those showing signs of previous treatment for conservation. Straight fork length (SFL) was estimated using an allometric equation [SFL = (2.2774 x weight) + 55.014; R2 = 0.975], using weight and length data of T. alalunga from our sampled fish and those available in Médieu et al. (2023). We used only average values, since raw original data from those studies were not available, and only fish sampled in the Atlantic Ocean were considered. The aim of this exercise is to easily compare with other studies when only length values were reported. Our allometric interpretations, however, are based on our 44 samples only. Muscle samples were stored in individually labelled plastic zip lock bags, frozen at a minimum of -20 ºC, and transported to the laboratory for analysis. The entire muscle samples were lyophilized over 48-72 h and the percentage of moisture gravimetrically estimated to convert Hg concentrations from dry to wet basis (UN Environment, 2019).

Samples of 0.5 g of lyophilized tissue were digested in 10 mL of concentrated nitric acid (HNO3, 65%) using heating and temperature ramp. 30 min at 200 °C. determined by MARS XPRESS - CEM microwave equipment. After digestion, 1 mL of hydrogen peroxide (H2O2, 35%) was added to the sample to avoid re-adsorption of Hg (Lacerda et al., 2017). Sample extracts were taken to 100 mL flasks with Milli-Q® water. The total Hg fraction obtained with this digestion procedure includes all inorganic and organic Hg species present in fish muscle tissue. All materials in contact with the samples were washed with acid and duplicate procedural blanks were included in each digestion bath. Subsequent Hg quantification was done by cold vapor atomic absorption spectrometry on a Nippon Instruments Corporation (NIC) model RA3210A. For the reduction of Hg we used a stannous chloride solution (SnCl2) prepared in 10% sulfuric acid (20% H2SO4). The estimated mean and standard deviation of the linearity coefficient of the calibration curves was 0.9998 ± 0.0001 and the mean and standard deviation of the detection limit was 0.03 ± 0.01 ng g−1 wet weight, calculated as three times the standard deviation of the reagent blanks divided by the slope of the calibration curve. Certified standards (NIST 2976 Mussel Tissue) were simultaneously analyzed with every analysis batch and presented a mean and standard deviation of Hg recovery of 95.0 ± 14.7% (n = 8). All concentrations commented are reported relative to wet weight (w.w.) obtained from our samples (FAO Codex Alimentarius Commission, 2021), whereas dry weight concentrations are listed in Table S.1.

Sub-samples of lyophilized tissue (200 mg) were weighed in PTFE tube and 5.0 mL of 25% KOH methanolic solution added to extract methyl-Hg in an oven at 70 ºC for 6 h with gentle stirring every hour. Sample ethylation (300 µL of 2 mol L^{-1} acetate buffer (pH 4.5) + 50 µL of tetra ethyl sodium borate (1%) for 30 µL of sample) followed Taylor et al. (2011) and a final volume brought to 40 mL with ultra-pure water (milli-Q, Millipore, Cambridge, MA, USA). Methyl-Hg quantification was performed on a gas chromatograph coupled to an atomic fluorescence spectrometer (GC-AFS - MERX-TM automated methyl-Hg system (Brooks Rand Labs. Seattle, USA). Reference material (Tuna Fish - BCR-463), run with each batch of samples, gave and average recovery of 96%, while detection (LOD) and quantification (LOQ) limits were 0.003 ng g-1 and 0.009 ng g-1 ww, respectively.

Sub-samples of lyophilized tissues (~ 1 mg) were weighed in tin capsules and analyzed in a Flash 2000 elemental analyzer coupled to a continuous flow mass spectrometer (Isotope Ratio Mass Spectrometry – IRMS, Delta V Advantage, Thermo Scientific, Germany) to quantify the stable isotopes. All results are expressed as delta value (δ), relative to Pee Dee Belemnite notation for $\delta^{13}C$ in parts per thousand (‰) and atmospheric N^2 for $\delta^{15}N$, according to (Fry, 2006). Analytical replicates showed variations lower than 5% and mean and standard deviation of the recovery of certified standard of protein (B2155) of 97 ± 1%.

Statistical analyses were performed using R 4.1.2 (R Development Core Team 2021). Data normality was tested using the Shapiro-Wilk test. All tests were conducted assuming a significance level of 95% ($p < 0.05$).

Individuals of T. alalunga analyzed showed a small variation of size with total weight varying from 20.8 to 44.6 kg (with mean and standard deviation of 28.5 ± 4.5 kg) and estimated straight fork length of 60 to 68 cm (with mean and standard variation of 62.9 ± 1.3 cm), slightly smaller than reported T. alalunga from other sites in the Atlantic. Total mercury concentrations (THg) varied from 294 ng g-1 to 930 ng g-1 ww, with a mean and standard variation of 515 ± 145 ng g^{-1} ww. The average percentage of methyl-Hg to total Hg concentrations was 92.0% (Table 1 and Table S.1).

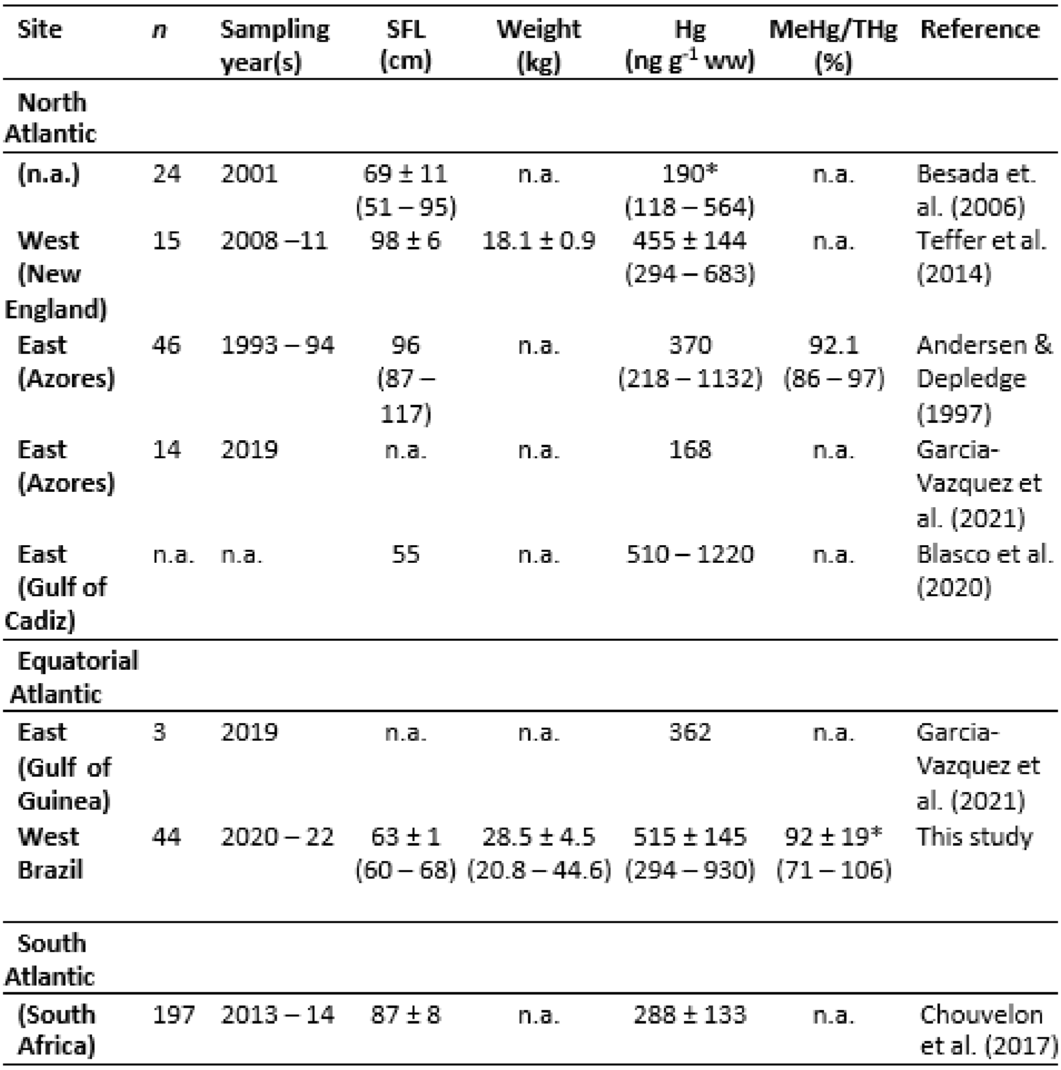
Table 1. Fish weight, stable isotope composition ($\delta^{13}C$ and $\delta^{15}N$), total Hg concentrations and percent of methyl-Hg, (mean ± standard deviation and range) of *Thunnus alalunga* sampled the Western Equatorial Atlantic Ocean (n = number of samples; SFL = estimated straight fork length based on weight data).

Fish Weight (kg)	**$\delta^{13}C$ (‰)**	**$\delta^{15}N$ (‰)**	**THg (ng g^{-1} ww)**	**Methyl-Hg/THg (%)**
28.5 ± 4.5 (20.8 – 44.6) n = 44	-17.8 ± 0.3 (-17.0 – -18.0) n = 42	11.3 ± 0.8 (9.8 – 13.3) n = 42	515 ± 145 (294 – 930) n = 44	92 ± 19 (71 – 106) n = 4

Although being smaller relative to individuals reported other sites in the Atlantic Ocean, the observed average Hg concentrations in individuals of *T. alalunga* from the Western Equatorial Atlantic are, in general, higher than in other Atlantic Ocean subregions, where Hg concentrations in this species have been reported. This is particularly true when only the Equatorial (Gulf of Guinea; Hg = 362 ng g^{-1} ww) (Garcia- Vazquez et al., 2021) and the Southeast Atlantic (South Africa; Hg = 288 ng g^{-1} ww) (Chouvelon et al., 2017) are considered (Table 2). When the ranges of concentrations are compared, however, higher maximum concentrations were reported in the Azores, by Andersen & Depledge (1997) (Hg = 1132 ng g^{-1} ww) and in the Gulf of Cadiz, by Blasco et al. (2020) (Hg = 1220 ng g^{-1} ww) (Table 2). Globally, Hg concentrations were similar to those compiled by Médieu et al. (2023) for T. alalunga in the Northwest (Japan) and Central (Hawaii) Pacific Ocean, and in the Indian Ocean but were lower than the reported concentrations in the Mediterranean. The fraction of MeHg relative to total is in general higher than most reported values but similar to those reported for albacore in the Mediterranean Sea (Storelli et al., 2002) and in the Azores (Atlantic Ocean) (Andersen & Depledge 1997).

This is the first report on THg concentrations in *T. alalunga* from the Western Atlantic, which makes difficult any within-region comparisons of Hg concentrations in this species. However, inter-species comparisons of Hg concentrations with other species of tuna (*T. albacares* (yellowfin) and *T. obesus* (Bigeye) caught in the Western Equatorial Atlantic (Lacerda et al., 2017), the same region of the present study showed higher average Hg concentrations in *T. obesus* (545; 95 - 1748 ng g^{-1}), followed by those in *T. alalunga* (515; 294 - 930 ng g^{-1}) and lower concentrations in *T. albacares* (159; 48 – 500 ng g^{-1}). This distribution of Hg concentrations is similar when the three species are compared on a global scale. This suggests that different biological and ecological traits (lifespan, growth rates, foraging depth and diet preferences) rather than geography explain the differences in THg concentrations among these three species, as previously suggested by Besada et al. (2006); Médieu et al. (2022; 2023) and Goyanna et al. (2023).

Table 2. Total Hg concentrations, percent of methyl-Hg and fish size (median* or mean ± standard deviation and range) of Thunnus alalunga sampled from different sites in the Atlantic Ocean (n = number of samples; SFL = straight fork length (estimated values from allometric relationship with total weight); n.a. = not available; *n = 4).



Site	*n*	Sampling year(s)	SFL (cm)	Weight (kg)	Hg (ng g⁻¹ ww)	MeHg/THg (%)	Reference
North Atlantic							
(n.a.)	24	2001	69 ± 11 (51 – 95)	n.a.	190* (118 – 564)	n.a.	Besada et. al. (2006)
West (New England)	15	2008 –11	98 ± 6	18.1 ± 0.9	455 ± 144 (294 – 683)	n.a.	Teffer et al. (2014)
East (Azores)	46	1993 – 94	96 (87 – 117)	n.a.	370 (218 – 1132)	92.1 (86 – 97)	Andersen & Depledge (1997)
East (Azores)	14	2019	n.a.	n.a.	168	n.a.	Garcia-Vazquez et al. (2021)
East (Gulf of Cadiz)	n.a.	n.a.	55	n.a.	510 – 1220	n.a.	Blasco et al. (2020)
Equatorial Atlantic							
East (Gulf of Guinea)	3	2019	n.a.	n.a.	362	n.a.	Garcia-Vazquez et al. (2021)
West Brazil	44	2020 – 22	63 ± 1 (60 – 68)	28.5 ± 4.5 (20.8 – 44.6)	515 ± 145 (294 – 930)	92 ± 19* (71 – 106)	This study
South Atlantic							
(South Africa)	197	2013 – 14	87 ± 8	n.a.	288 ± 133	n.a.	Chouvelon et al. (2017)

Stable isotope composition (δ^{13}C: -17.0 ‰ to -18.0 ‰ and δ^{15}N: 9.8 ‰ to 13.28 ‰) (Table 1) characterizes *T. alalunga* as an oceanic tertiary consumer and reflects a mainly piscivorous diet composed of fishes of the Scomberesocidae, Engraulidae, and Gadidae families (Duffy et al., 2017). Similarly, other tuna species, such as *T. obesus* and *T. albacares* (Pethybridge et al., 2018), also present isotope values corresponding to oceanic tertiary consumers with diet including prey from these and other fish families (Duffy et al., 2017). In contrast, Hg levels in *T. obesus* and *T. albacares* have been shown to differ mostly due to differences in preferred prey species, and foraging behavior/depth (Lacerda et al., 2017; Silva et al., 2019; Mesquita et al., 2021). Therefore, Hg levels can differ among species that feed in the same trophic levels reflecting species-specific foraging strategies. For *T. alalunga* in the present study, we found that stable isotope values are in the same range of those reported in other oceanic areas, which typical display high within-area variability, therefore direct comparisons may not be appropriate due to differences in baseline stable isotope values (Lorrain et al., 2020; Pethybridge et al., 2018).

The high fraction of Methyl-Hg to the total Hg concentrations (Tab. 1) also reflects a high trophic position and is similar to *T. albacares* (Voegborlo et al., 2006) and *T. obesus* (Médieu et al., 2022; 2023). However, the association of high %MeHg (92%) measured in the sampled albacore with its trophic position as a top predator, must be viewed with caution since low %MeHg has been reported in top predator tunas (Manceau et al.,2021).

There is a significant and positive linear correlation between fish weight and Hg concentration (Hg = 25 kg –190; r^2= 0.596; $p < 0.01$; n = 44) (Figure 2) as has been observed for most species of tuna, most likely, displaying a linear relationship with body dimensions (Drevnick et al., 2015). Most available data for his species in the Atlantic Ocean relates Hg concentrations with fish length. For example, Besada et al. (2006) reported a similar linear relationship between Hg concentrations and body length in larger individuals of *T. alalunga* from the North Atlantic (Hg = 65 cm –229; r^2 = 0.6). for comparison, we have transformed our weight (kg) data into length (cm) and obtained the equation: Hg = 57.2 cm – 3,074; r^2= 0.445; $p < 0.01$; n = 44.

The North Atlantic individuals with much larger size (51 to 95 cm) than those from this study (estimated body length varying from 60 to 68 cm), displays a much slower Hg accumulation than *T. alalunga* from the Equatorial Western Atlantic. On the other hand, in the Pacific Ocean, smaller individuals of *T. alalunga* also displays much faster Hg accumulation rates relative to body size, than larger individuals (Houssard et al., 2019).

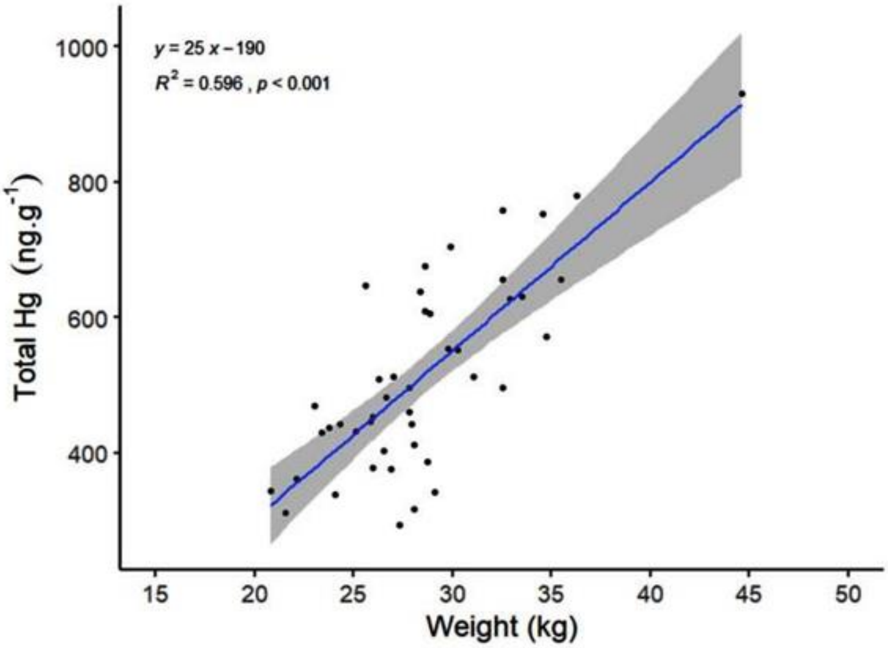


Figure 2. Relationship between total Hg concentrations and fish weight in T. alalunga caught in the Equatorial Southwestern Atlantic.

The concentrations of Hg observed in Albacore are generally safe for consumption as none of them exceeded the legal Hg limits (1000 ng g^{-1}) established to protect human consumers according to the Brazilian legislation. However, a recent estimate for those consuming fish on a regular and sustained basis (Bezerra et al., 2023), suggested some risk of long-term chronic exposure under concentrations below the legal limit (Fish Screening Level (FSL) of 300 mg kg^{-1}). The FSL represents an upper limit for Hg concentrations in fish and considers the reference dose for Hg (0.0001 mg kg^{-1} day^{-1}), the local fish consumption rate, and the consumer body weight. Thus, this parameter is more protective to fish consumers than the general guideline of 1 mg kg^{-1}. Considering the FSL level, Hg concentrations in Albacore in the present study were above this safety limit and, thus, it is advisable to exercise caution when consuming this species. In addition, a recent modelling of the global MeHg catches through fisheries (Wu & Zhang, 2023) highlighted the significance of Brazilian fisheries, with a significant fraction (~33%) of the Brazilian contribution to the global MeHg catches coming from the Western Equatorial Atlantic.

Albacore showed differences in Hg concentrations among the three major populations in the Atlantic Ocean and the Mediterranean Sea. Regional differences on Hg concentrations between the two sides of the Atlantic Ocean, show that the western population presents a higher

Hg content. These higher concentrations may be associated with different Methyl-Hg production rates and emission magnitude. Unfortunately, only a few studies exist on Hg distribution in tuna from the South Atlantic Ocean hampering further comparisons at this stage and limiting the use of *T. alalunga* as biomonitores of oceanic Hg concentrations. In addition, three is scarce, if any, data on the vertical and horizontal variability of Methyl-Hg in this ocean. Although Hg concentrations in *T. alalunga* from the Western Equatorial Atlantic are relatively high, they are still lower than the Brazilian limits for human consumption. However, local consumption rates significantly increase the risk of Hg exposure in humans especially when combining with the percentage of Methyl-Hg observed in this study, warrants the implementation of health control. As a recommendation following USEPA (2000) guidelines, we estimated a maximum consumption of four meals per month for the general population (men age 18+ and women age 46+), and three meals per month for children (< 12 years old) and women at reproductive age (18 to 45 years old), assuming a portion of 100 g and 50 g of fresh fish for adults and children, respectively.

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CAPÍTULO III: AVALIAÇÃO MULTIFATORIAL UTILIZANDO MERCÚRIO (Hg), ISÓTOPOS ESTÁVEIS ($\delta^{15}N$ & $\delta^{13}C$) E RECOMENDAÇÕES DE SEGURANÇA ALIMENTAR PARA TUBARÕES E PEIXES ÓSSEOS DE NÍVEL TRÓFICO SUPERIOR DO OCEANO ATLÂNTICO[3]

Resumo: O presente artigo apresenta dados inéditos de concentrações de mercúrio (Hg), isótopos estáveis ($\delta^{15}N$ & $\delta^{13}C$) de tubarões e peixes ósseos de alto nível trófico capturados no oceano Atlântico Equatorial Oeste e apresenta recomendações para o consumo seguro pela população. Encontramos diferenças significativas nas concentrações de Hg entre as espécies, verificando concentrações de Hg mais elevadas em *I. oxyrinchus*, *X. gladius* e *P. glauca*, enquanto as concentrações menores foram observadas em *T. albacares e C. hippurus*. Concentrações intermediárias de Hg foram observadas em *T. alalunga*, *I. albicans* e *T. obesus*. A proporção de metilmercúrio em relação ao Hg total quantificado em quatro espécies foi de 82,9 ± 5,3% em *C. hippurus*, 82,0 ± 21,4% em P. glauca, 92,0 ± 15,2% em *T. alalunga* e 82,1 ± 1,9% em *X. gladius*. Os valores de $\delta^{15}N$ variaram de 10,21 ± 0,37‰, em *T. albacares* a 12,39 ± 1,1‰ em *X. gladius*. Os maiores valores foram observados em *I. oxyrinchus*, *X. gladius* e *P. glauca*, em comparação com *C. hippurus*, *I. albicans* e *T. albacares*, que apresentaram os menores valores de $\delta^{15}N$. Os valores de $\delta^{13}C$ variaram de -18,40 ± 2,6 ‰ em *X. gladius* a -16,62 ± 0,4‰ em *C. hippurus*, com diferenças significativas entre as espécies. Foi verificada correlação significativa entre as concentrações de Hg e o peso dos peixes para todas as espécies, exceto *C. hippurus, I. albicans* e *I. oxyrinchus*. O teste de correlação entre as concentrações de Hg transformadas em log e $\delta^{15}N$ foi significativo apenas para *X. gladius* O teste de correlação entre as concentrações de Hg transformadas em log e $\delta^{13}C$ só foi significativo para o *C. hippurus*. Finalmente, foi estimada a dose mensal de consumo seguro para as populações que variou de 0 a 11. Foi maior em *T. albacares* e *C. hippurus*, porém para as espécies *X. gladius* e *I. oxyrinchus*, o número estimado foi zero para os consumidores infantis.

Palavras-chave: mercúrio, tubarões, Atlântico, atum, isótopos, contaminação

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Multifactorial evaluation using mercury (Hg), stable isotopes ($\delta^{15}N$ and $\delta^{13}C$) and food safety recommendations for upper trophic level sharks and pelagic fishes.

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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 recommendations of safe consumption by human populations. We found significant differences in Hg concentrations among species, with highest Hg concentrations in *Isurus oxyrinchus* followed by *Xiphias gladius* and *Prionace glauca*, while lowest Hg concentrations were observed in *Thunnus albacares* and *Coryphaena hippurus*. Intermediate Hg concentrations were observed in *T. alalunga*, *Istiophorus. albicans* and *T. obesus*. 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 ranged from 10.21 ± 0.37‰ in *T. albacares* to 12.39 ± 1.1‰ in *X. gladius*. The highest values were observed in *I. oxyrinchus*, *X. gladius* and *P. glauca*, compared to *C. hippurus*, *I. albicans* and *T. albacares*, which showed the lowest $\delta^{15}N$ values. The $\delta^{13}C$ values ranged from -18.40 ± 2.6 ‰ in *X. gladius* to -16.62 ± 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 $\delta^{15}N$ indicate this pollutant is biomagnifying in the upper trophic levels of this oceanic trophic web. The estimated species-specific number of meals that is safe for consumption ranged from 0 to 11 meals per month for adults and children.

Keywords: methylmercury, sharks, Atlantic, tuna, isotopes, contamination

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 (CH_3Hg) (Obrist et al., 2018). The methylmercury present in the marine ecosystem can be assimilated in the food web, through direct uptake from the water in phytoplankton, and be transferred to other organisms through feeding (Gosnell et al., 2021). Less efficient assimilations include absorption by fish gills, and deposition during the synthesis of invertebrates' hard structures (Saidon et al., 2024). Characteristics such as body size, age, habitat, trophic position, and life span are known to control the magnitude of Hg accumulation, while physiological characteristics control Hg distribution in fish tissues and organs (Schneider et al., 2013). Hg biomagnifies in aquatic food webs resulting in upper-level predators presenting concentrations several times higher than primary producers. 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 highlight the importance of evaluating multiple species in biomonitoring efforts (Médieu et al., 2023).

Long-term mercury monitoring in these fish is also limited and current information on Hg accumulation shows greater than expected levels considering changes in global atmospheric emissions and deposition (Médieu et al., 2023). The distribution of mercury (Hg) on a global scale in large oceanic carnivorous fish, mainly tuna, tuna-like and sharks species have shown that variations in mercury levels are partly driven by species-specific ecological characteristics, such as foraging depth, diet composition and body size (Lacerda et al., 2017), which suggest that large bony fish and sharks can provide complementary information on the vertical and horizontal distribution of methylmercury in the ocean (Goyanna et al., 2023; Médieu et al., 2023).Therefore, 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 disentangle these from the spatial variability in Hg concentrations, both vertically through the water column and horizontally between oceanic regions. Furthermore, there is still significant gaps in Hg accumulation data for many pelagic species from oceanic regions in the South Atlantic Ocean (Goyanna et al., 2023; Médieu et al., 2023).

Ecological drivers often described to influence Hg accumulation in marine megafauna are 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). All 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 most commonly used to examine various trophic processes (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 the 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 fish with higher trophic levels. 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 quantifying the trophic biomagnification factor (TMF) of mercury in the trophic web, providing information on the magnitude of Hg increases across multiple trophic levels (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, which is larger than aquaculture (inland and marine) and inland captures. Provisional estimates suggest that 88.4% of the average production is intended for human consumption, resulting in a per capita apparent consumption of 20.4 kg per year (or 55.9 grams per day) (FAO, 2022).

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). 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 in 2021 per species type were 4,629 t for the Blue Shark, *Prionace glauca*, 2,110 t for the Swordfish, *Xiphias gladius*, 6.499 t for the bigeye, *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).

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 fish species (Goyanna et al., 2023). Furthermore, Hg levels in these species are generally in the organic, and most toxic, form of methylmercury (> 80%) (Monteiro and Lopes, 1990; Storelli and Marcotrigiano, 2001). It has been estimated that upper trophic level fish from the Brazilian Equatorial Atlantic Coast, including the Amazon River discharge area, are among the highest in methylmercury concentration globally (Wu and Zhang, 2023). As a result, despite of Brazil accounting for less than 1% of the global fish catches (FAO, 2022), its estimated contribution to methylmercury exports through fish catch accounts for 21% of total. (Wu and Zhang, 2023).

Therefore, the goal of the present study is three-fold 1) To assess Hg accumulation in upper trophic level pelagic fish in the Equatorial Western Atlantic Ocean, including the estimation of trophic magnification factor (TMF); 2) To evaluate ecological drivers of Hg accumulation using $\delta^{15}N$ and $\delta^{13}C$; and 3) To estimate human exposure to Hg through fish consumption while providing consumer-friendly recommendations of the best choices of consumption. Specifically, we ask the following research questions to guide our study: 1) Do Hg accumulation levels in upper trophic level fish differs across species and geographical location? Considering previously reported global Hg data in tuna and tuna-like species, we hypothesize that Hg levels in upper trophic level pelagic fish from the Equatorial Western Atlantic differ among species and are lower compared to similar species occurring in other oceanic regions. 2) Does species-specific ecological characteristics (e.g. isotopic niche) explain Hg accumulation in upper trophic level pelagic fish? Considering 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) Does upper trophic level pelagic fish accumulate Hg at levels potentially harmful to human consumers? Considering the high trophic position of these fish and the known biomagnification of Hg in marine food-webs, we hypothesize Hg in these large pelagic fish are in potentially harmful levels to human consumers.

Material and Methods

Sampling description

Muscle samples of *Coryphaena hippurus, I. albicans, I. oxyrinchus, P. glauca, T. alalunga* and *X. gladius* studied were obtained directly from the fishing industry, during the landings of duly licensed vessels operating with pelagic longlines in Natal, Rio Grande do Norte state (RN), in Brazil, during two seasons (August 2020 and October 2022). These species were caught at geographical coordinates between 3°N - 6°S and 16°- 30° W (Figure 1).

The samples of *T. obesus* and *T. albacares* were obtained directly from the tuna fleet in the associated shoal modality, based in the port of Areia Branca, RN, during two distinct seasons (July 2014 and October 2015). This tuna fleet typically operates around the buoy of the PIRATA Program (Pilot Moored Array in the Tropical Atlantic), anchored at position 0º N; 35º W, at an approximate distance of between 320 and 380 nautical miles from the coast, with depths of approximately 4,000 m. Capture techniques employed include trolling, hand line, and pole and line (Silva et al., 2016).

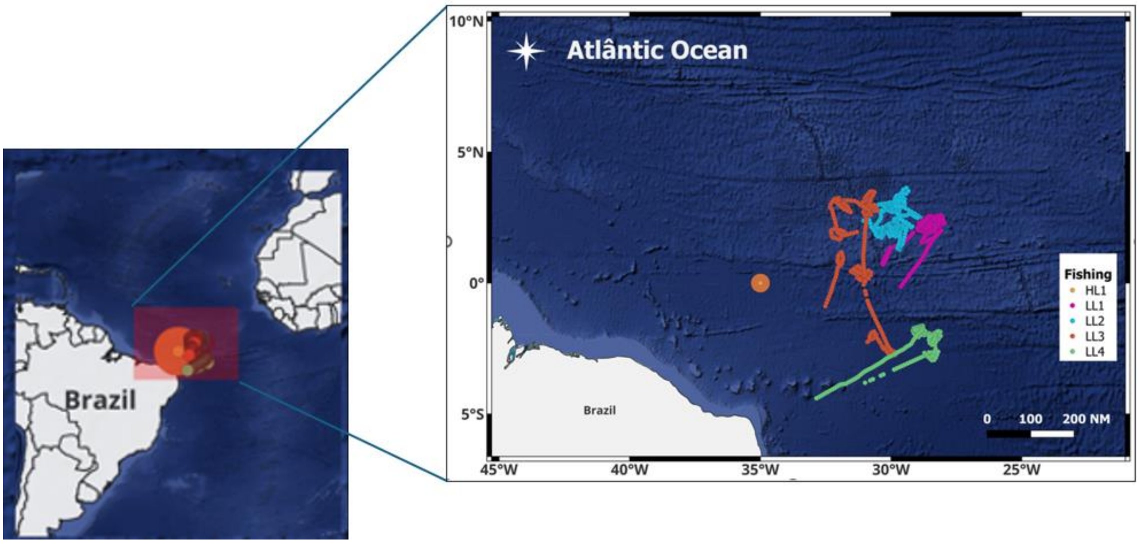

Figure. 1 - Map of the Atlantic presenting the fishing areas from both pelagic longlines (LL) and aggregated schools (HL).

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 to 96 hours 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.

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). The samples were weighed in duplicate to a weight of approximately 0.5 g in Teflon tubes, where 10 mL of concentrated nitric acid (HNO_3 65%) was added for a pre-digestion period of 25 min. After the pre-digestion, the digestion was carried out in a microwave oven with 1600 W power and a temperature of 200°C for 30 min. Then 1 mL of hydrogen peroxide (H_2O_2) was added. The final extract was transferred to 100 mL volumetric flasks and filled with distilled water. For each analysis, the materials used were washed in a bath of neutral detergent followed by HCl 10%. The calibration was performed using a stock solution (MERCK) containing 1,000 $\mu g. mL^{-1}$ of Hg. Successive dilutions were made until a working solution of 1 $ng. mL^{-1}$ of Hg was obtained. This solution was used to draw up the calibration curves.

For methyl mercury (MeHg) determination, about 100mg (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 6h, 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 $NaBEt_4$ 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).

For every total Hg analysis batch (n = 26), the methodology was validated using a certified reference material (SRM), which was fish muscle tissue (ERMBB422) containing 601 ± 30 ng g^{-1} for THg and MeHg (BCR-463) containing 304 ± 16 ng g^{-1} were used. The average value obtained is shown in the table 1 below:

Table 1 - Total THg and MeHg concentrations in certified standard and values obtained and respective recovery.

SRM	Certified Value (ng.g^{-1})	Value obtained (ng.g^{-1})	Recovery (%)
ERM-BB422	601 ± 30	660 ± 99	≥90
BCR-463	304 ± 16	265 ± 21	≥87

Stable Isotope Analysis

After lyophilization, muscle tissue samples weighing about 0.5 mg (dry weight) were placed in tin capsules. The elemental and isotope ratios of carbon ($^{13}C/^{12}C$) and nitrogen ($^{15}N/^{14}N$) were quantified in an isotopic mass spectrometer (Thermo Finnigan Delta V Advantage) coupled to an organic elemental analyzer (Flash 2000 - Thermo Fisher Scientific). The results have a reproducibility and accuracy of 95% measured using replicates (n=3) and a certified standard (Protein cat. B2155).

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 Equation (1).

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

where BW is the average consumer body weight in kilograms, CR is the daily fish consumption rate (in kilograms per day), and RfD is the reference dose of 0.1 mg kg bw −1 day−1. RfD indicates the daily exposure estimated to pose no risk of harmful health effects over a lifetime. We estimated FSL parameter for two fish consumption scenarios, 1) A CR of 24.5 grams per day reflecting the Brazilian per capita fish consumption for northeastern Brazilian population (e.g. general) (IBGE, 2021), and 2) A CR of 142 grams per day reflecting fish intake of populations that eat fish daily or as subsistence. We also estimated the Hg exposure for two consumer populations, 1) Adult 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) to solve for daily fish consumption rate (CR), 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 Equation (2):

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

where T_{ap} is the averaging time period (365.25 days per 12 months or 30.44 days per month), and MS represents the average meal size (in kilograms per meal) (USEPA, 2000). For our calculations, we assumed an average fish meal size of 150 grams for adults (BRASIL, 2014) and 75 grams for children.

We estimated the daily intake (EDI_{Hg} mg kg_{BW}^{-1} day^{-1}) for both consumption scenarios and all consumer groups using equation (3)

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

Finally, we evaluated the target hazard quotient (THQ) using Equation (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 X ED).

Statistical analysis

All statistical analyses and plotting figures were performed using R version 3.4.3 (R Core Team, 2017). 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 a post hoc pairwise Wilcoxon Rank tests, with Holm corrections, to test for differences in Hg accumulation and $\delta^{13}C$ values among species. We conducted parametric ANOVA followed by post-hoc Tukey test to test differences in $\delta^{15}N$. We used Pearson's correlation to check the relationship between Hg and weight, and Sperman's correlation to check the relationship between Hg and other variables. Statistical significance was set at $p = 0.05$ for all tests.

Results

We collected 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). That is because we used stored muscle samples from the same individuals to quantify additional variables, such as $\delta^{13}C$, $\delta^{15}N$, and methylmercury, and thus conducting a complete assessment of factors affecting Hg accumulation.

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 2). The smallest individual sampled was a *T. albacares* weighing 1.3 kg, and the largest was a *X. gladius* weighing 127.1 kg (Table 2).

Mercury accumulation description

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 studied species. The average proportion relative to total Hg quantified 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 total Hg is in the organic methylmercury form. It should be noted that the MeHg values refer only to the fish that were analyzed for this parameter and therefore do not reflect all the samples that were analyzed for THg.

Median THg concentrations were highest in *I. oxyrinchus* (2,152.8 ± 1,110.9 ng g^{-1}), followed by *X. gladius* (1,414.6 ± 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 Hg 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 2). The lowest THg concentration was observed in an *I. albicans* (75.3 ng g^{-1}) individual, and the highest in an *I. oxyrinchus* (5,159.6 ng g $^{-1}$) individual (Table 2). We found significant differences in THg concentrations among species (Kruskal-Wallis H = 197.0; df = 7; n = 252; $p < 0.001$) (Figure 2).

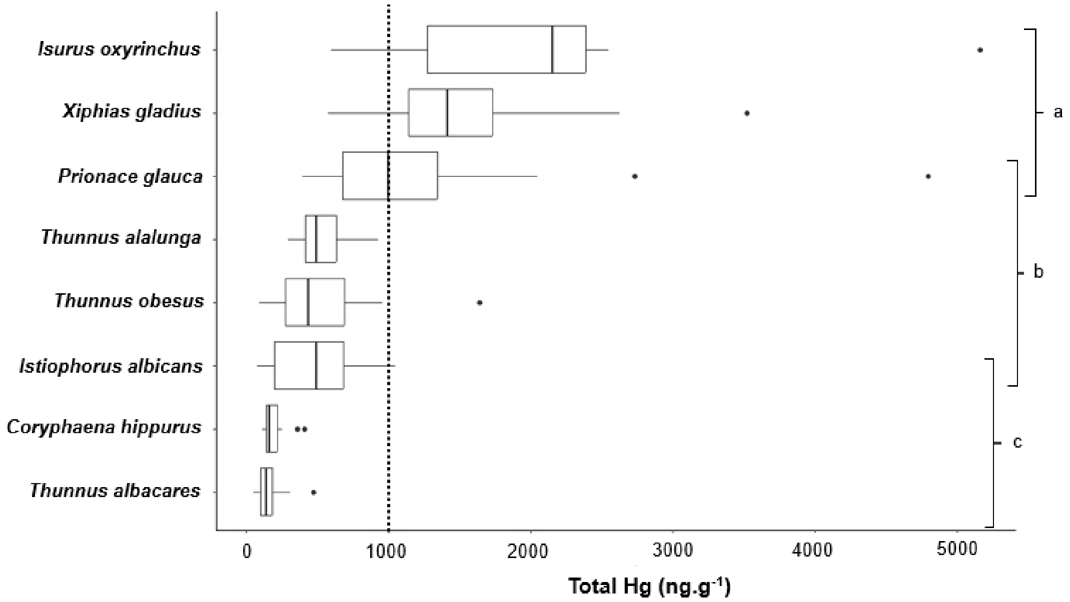

Figure 2 - Boxplot with Hg variations in 8 species of oceanic fish from western equatorial Atlantic. Letters a, b and c represent statistically significant differences ($p > 0.05$). The dashed line indicates predator fish maximum limit for THg concentration stablished by the regulatory agencies.

Table 2 - Descriptive table with the mean, standard deviation, minimum and maximum values for weight, mercury (THg) and methylmercury (MeHg) of the species studied.

Species (n)	Weight (kg)				THg (ng.g^{-1} w.w.)				MeHg (ng.g^{-1} w.w.)			
	Mean	SD	Min	Max	Median	SD	Min	Max	Mean	SD	Min	Max
Coryphaena hippurus (17; 4)*	9.67	7.39	4.07	35.97	157.70	82.80	111.65	407.30	288.76	155.27	134.73	482.81
Istiophorus albicans (8)	26.65	8.70	13.00	38.48	487.20	348.57	75.28	1048.70				
Isurus oxyrinchus (7)	60.90	13.54	40.46	77.00	2152.80	1521.51	593.14	5159.60				
Prionace glauca (50, 29)*	39.28	11.57	16.80	70.00	998.60	830.75	453.23	4792.00	1001.10	476.94	240.02	2370.52
Thunnus alalunga (44; 4)*	28.52	4.50	20.80	44.59	488.90	145.96	294.46	930.10	561.88	190.89	342.63	691.21
Thunnus albacares (52)	16.76	13.41	1.31	60.00	136.50	42.61	125.70	236.40				
Thunnus obesus (30)	13.07	9.03	2.92	51.02	432.30	343.09	95.40	1748.10				
Xiphias gladius (44; 18)*	55.50	19.11	32.75	127.07	1414.60	671.89	646.78	3521.20	1414.97	691.38	657.12	3606.18

*denotes the sample number of methylmercury analysis.

Description of stable isotope values (δ^{15}N and δ^{13}C)

We determined stable isotope ratios for a subset of collected species 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). Estimated isotopic niche for the eight upper trophic level species is shown in Figure 3.

The average δ^{15}N values ranged from 10.21 ± 0.37‰, in *T. albacares*, to 12.4 ± 1.1‰ in *X. gladius* (Table 3). Significant differences among species were observed for δ^{15}N values (ANOVA $F_{7,125} = 10.76$; $p < 0.001$) (Figure 4). 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 δ^{15}N. Intermediate values were observed in *T. obesus* and *T. alalunga* (Figure 4).

The δ^{13}C values ranged from -18.4 ± 2.6 ‰, in *X. gladius*, to -16.6 ± 0.4‰ in *C. hippurus* (Table 3). Significant differences among species were observed for δ^{13}C values (Kruskal-Wallis $H = 44.7$; $df = 7$, $n = 133$, $p < 0.001$) (Figure 4). 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 (Figure. 4).

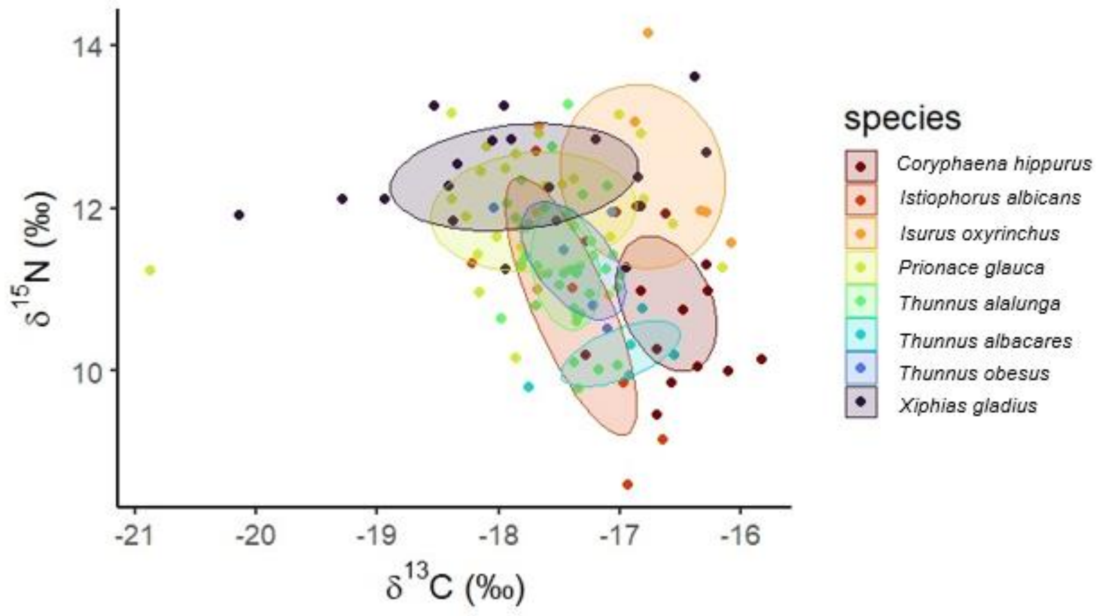

Figure 3 – Isotopic niche occupied by eight fish species of higher trophic level.

Table 3 – Descriptive table with the median, 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	Median	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	-17.5	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	-16.9	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	-17.2	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	-18.0	2.6	-28.5	-16.3

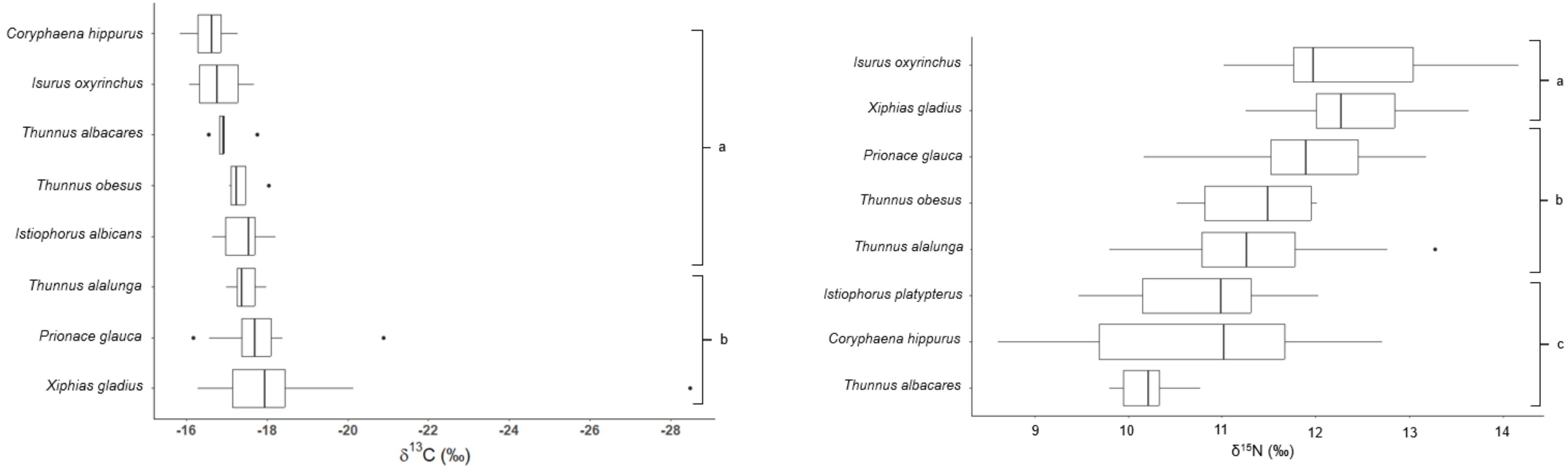

Figure 4 – Boxplot with $\delta^{13}C$ and $\delta^{15}N$ variations in eight species of oceanic fish from Equatorial Atlantic. Letters a, b and c represent statistically differences ($p > 0.05$).

Correlations between Hg, weight and isotopes

We observed a significant correlation between Hg concentrations and fish weight for all species (Figure 5; Table 4), 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 δ^{15}N values (Figure 6). However, when performed on each species separately, correlation test between Log-transformed Hg concentrations and δ^{15}N was significant for *X. gladius* only (Pearson, r = 0.54, df = 18, p = 0.012) (Table 4). Correlation test between Log-transformed Hg concentrations and δ^{13}C was significant for *C. hippurus* only (Pearson, r = 0.59, df = 15, p = 0.01) (Table 4).

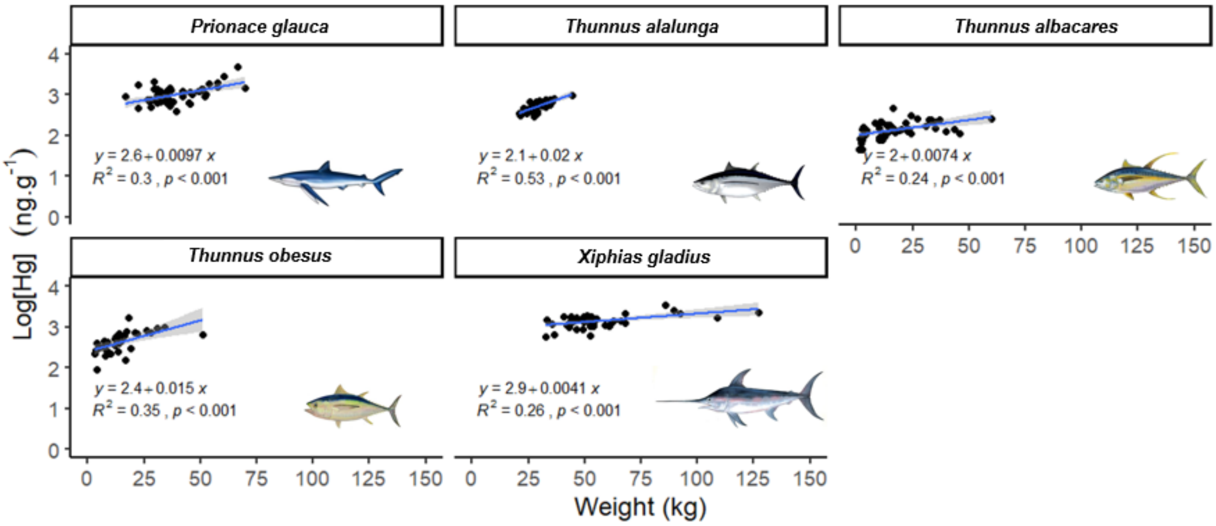

Figure 5 - Correlation between Hg concentrations and fish weight for species with a positive correlation.

Table 4 – Results of Spearman or Pearson correlation test between Hg concentrations and other variables (weight, δ^{15}N and δ^{13}C).

	Fish Weight (kg)			δ^{15}N (‰)			δ^{13}C (‰)		
	df	r	p	df	r	p	df	r	p
Coryphaena hippurus	15	0.18	0.47	15	-0.34	0.17	15	0.59*	0.01
Istiophorus albicans	6	0.52*	0.18	6	0.45	0.26	6	-0.62	0.09
Isurus oxyrinchus	5	0.25*	0.58	5	-0.17	0.71	5	-0.42	0.35
Prionace glauca	48	0.54*	<0.001	27	-0.06	0.75	27	0.06	0.72

Thunnus alalunga	42	0.69	<0.001	40	-0.32*	0.04	40	0.22*	0.16
Thunnus albacares	45	0.56	<0.001	3	-0.08*	0.89	3	0.26*	0.67
Thunnus obesus	28	0.71	<0.001	3	0.07*	0.91	3	0.31*	0.61
Xiphias gladius	42	0.32	0.03	18	0.54*	0.012	18	0.09*	0.70

*Denotes Pearson correlation coefficient.

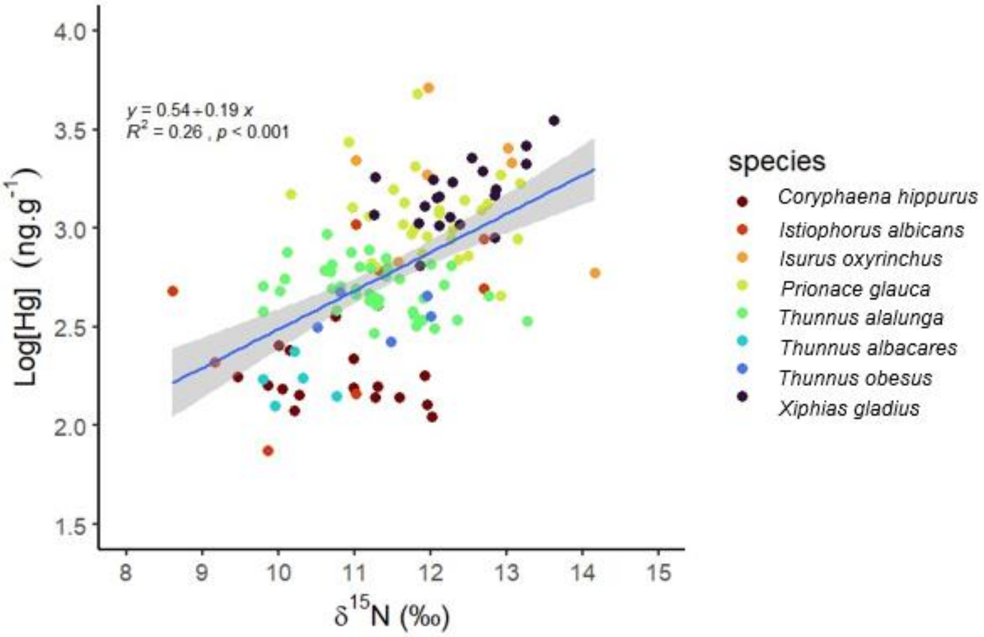

Figure 6 - Relationship between Log-transformed Hg concentrations and δ^{15}N of the species.

Risk exposure and intake 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 adults and children under two consumption scenarios, general intake (CR = 24.5 g.day^{-1}) and subsistence intake (CR = 142 g.day^{-1}) (Table 5 and 6).

We found that THQ values estimated for adult general consumers were above 1 (indicating potentially excessive exposure) for the majority of species, except for *C. hippurus* and *T. albacares*. Similarly, THQ values estimated for adults and children in a 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 ranged 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 5).

Using mean values of THg concentration, we estimated the safe amount of ingested fish and subsequent number of meals per month for each species (Table 6). Maximum consumption rate (CR_{max}) mean values varied for adults and children in a general or subsistence consumption scenario. Estimated safe number of meals (CR_{mm}) varied from 0 to 11 meals per month, and was highest in *T. albacares* and *C. hippurus*. For the species *X. gladius* and *I. oxyrinchus* the estimated safe number of meals was below one meal per month for children consumers.

Table 5 - Mean (standard deviation) of EDI (mg $kg^{1}{}_{BW}$ day^{-1}) in all scenarios. The reference dose (RfD) for Hg is 0.0001 mg $kg^{-1}{}_{BW}$ day^{-1}. ag = adults general, cg = children general, as = adult subsistence and cs = children subsistence

Species (n)	THg	EDI ag	EDI cg	EDI as	EDI cs
	Mean ± SD	Mean ± SD	Mean ± SD	Mean ± SD	Mean ± SD
Coryphaena hippurus (17)	190.70 ± 82.80	0.0001 ± 0.0000	0.0003 ± 0.0001	0.0004 ± 0.0002	0.0018 ± 0.0008
Istiophorus albicans (8)	493.10 ± 348.57	0.0002 ± 0.0001	0.0008 ± 0.0006	0.0010 ± 0.0007	0.0047 ± 0.0033
Isurus oxyrinchus (7)	2173.60 ± 1521.51	0.0008 ± 0.0005	0.0036 ± 0.0025	0.0044 ± 0.0031	0.0206 ± 0.0144
Prionace glauca (50)	1132.60 ± 830.75	0.0004 ± 0.0002	0.0018 ± 0.0011	0.0023 ± 0.0014	0.0107 ± 0.0066
Thunnus alalunga (44)	514.70 ± 145.96	0.0002 ± 0.0001	0.0008 ± 0.0002	0.0010 ± 0.0003	0.0049 ± 0.0014
Thunnus albacares (52)	149.40 ± 42.61	0.0001 ± 0.0000	0,0002 ± 0.0001	0.0003 ± 0.0002	0.0015 ± 0.0007
Thunnus obesus (30)	511.60 ± 343.09	0.0002 ± 0.0001	0,0009 ± 0.0006	0.0011 ± 0.0007	0.0050 ± 0.0032
Xiphias gladius (44)	1468.80 ± 671.89	0.0005 ± 0.0002	0.0024 ± 0.0009	0.0030 ± 0.0011	0.0139 ± 0.0051

Table 6 - Mean (standard deviation) of risk variables and intake recommendations. CR$_{max}$ is expressed in g.day^{-1} and CR$_{mm}$ in meals per month. ag = adults general, cg = children general, as = adult subsistence and cs = children subsistence

Species (n)	THg	THQ ag	THQ cg	THQ as	THQ cs	CR$_{max}$ Adult	CR$_{max}$ Child	CR$_{mm}$ Adult	CR$_{mm}$ Child
	Mean ± SD	Mean ± SD	Mean ± SD	Mean ± SD	Mean ± SD	Mean ± SD	Mean ± SD	Mean ± SD	Mean ± SD
Coryphaena hippurus (17	190.70 ± 82.80	0.67 ± 0.29	3.12 ± 1.35	3.87 ± 1.68	18.06 ± 7.84	42 ± 13	9 ± 3	8 ± 3	4 ± 1
Istiophorus albicans (8)	493.10 ± 348.57	1.73 ± 1.22	8.05 ± 5.69	10.00 ± 7.07	46.68 ± 33.00	29 ± 30	6 ± 6	6 ± 6	2 ± 3
Isurus oxyrinchus (7)	2173.60 ± 1521.51	7.61 ± 5.3	35.50 ± 24.85	44.09 ± 30.86	205.77 ± 144.04	5 ± 4	1 ± 1	1 ± 1	0 ± 0
Prionace glauca (50)	1132.60 ± 830.75	3.96 ± 2.5	18.50 ± 11.47	22.98 ± 14.25	107.22 ± 66.50	8 ± 3	2 ± 1	2 ± 1	1 ± 0
Thunnus alalunga (44)	514.70 ± 145.96	1.80 ± 51	8.41 ± 2.37	10.44 ± 2.95	48.73 ± 13.75	15 ± 4	3 ± 1	3 ± 1	1 ± 0
Thunnus albacares (52)	149.40 ± 42.61	0.56 ± 0.28	2.62 ± 1.29	3.25 ± 1.60	15.16 ± 7.48	54 ± 27	12 ± 6	11 ± 5	5 ± 2
Thunnus obesus (30)	511.60 ± 343.09	1.86 ± 1.20	8.68 ± 5.60	10.79 ± 6.96	50.34 ± 32.48	19 ± 14	4 ± 3	4 ± 3	2 ± 1
Xiphias gladius (44)	1468.80 ± 671.89	5.14 ± 1.88	23.99 ± 8.78	29.79 ± 10.91	139.04 ± 50.91	5 ± 2	1 ± 0	1 ± 0	0 ± 0

Discussion

Ecological characteristics and mercury accumulation in species.

Mercury (Hg) concentrations in are generally lower in juvenile individuals compared to adults, although this relationship is highly variable (Goyanna et al. 2023). Ontogenetic changes that occur when juvenile individuals acquire endothermic adaptations result in foraging at deeper waters where the Hg content of the prey is higher due to intensified methylation processes and the carnivorous nature of deep-water trophic webs (Lamborg et al., 2014; Graham et al., 2007). Above a certain size, regardless of the species, Hg concentrations are more likely to be related to body size (Drevnick and Brooks, 2017). Corroborating this, previous literature suggests that oceanic pelagic fish from the South Atlantic tend to have higher mercury concentrations by weight and/or size compared to those from the North Atlantic (Goyanna et al., 2023; Lacerda et al., 2017). Other research has linked geographical variations to the extent of mercury emissions, as indicated by differing mercury concentrations in the North and South Atlantic Oceans (Lamborg et al. 2014).

In addition, variability in mercury concentrations among sharks, tunas and tunas-like species has been documented, with sharks showing higher concentrations, probably due to their intrinsic ecology characteristics of elasmobranchs (Goyanna et al., 2023). Even when subject to similar diets and habitats, elasmobranchs exhibit higher Hg concentrations than top predatory bony fish, indicating that elasmobranchs possess specific characteristics that promote Hg accumulation. These characteristics include slow growth, late maturation, low reproductive rates, and longevity, as well as unique metabolic and physiological processes (Dulvy et al., 2017, 2008). These factors make them particularly prone to accumulating high levels of contaminants and especially vulnerable to pollutants placing them at potential risk of negative health effects (Dulvy et al., 2017, 2008; Storelli et al., 2002; Tiktak et al., 2020).

P. glauca is a highly migratory species, with movement patterns associated with reproductive behavior and prey distribution (Nakano and Stevens, 2008). This species moves vertically in the water column, with depth distribution of 400 m to 1,000 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 (Bigelow et al., 1999), being most commonly found between latitudes 20º - 40ºS (Carvalho et al., 2011). Catches are more efficient where the water surface temperature is around 16-28ºC (Montealegre-Quijano and Vooren, 2010). The Hg levels in the present study are comparable to previous studies on the western south Atlantic Ocean with *P. glauca* of similar weights (Carvalho et al., 2014). In comparison to North Atlantic populations, studies have shown that larger individuals (114.2 ± 11.1 kg) (Hauser-Davis et al., 2021) accumulate Hg levels similar to those found in the present study, suggesting that *P. glauca* in the South Atlantic accumulate Hg to a greater extent. Compared to studies in the Pacific Ocean, Hg values are similar to those in the present study (Escobar-Sanchez et al., 2011; Barrera-García et al., 2012).

In contrast, for the *I. oxyrinchus* species, individuals captured in the North Atlantic Ocean show higher Hg concentrations (2,647 $ng.g^{-1}$, range 755-4,933 $ng.g^{-1}$) than those in the present study, but these individuals are slightly larger considering weigh values (82.6 ± 4.6 kg) (Teffer et al., 2014). High Hg concentrations (2,110 ng.g-1, range 590-5,580 $ng.g^{-1}$) were also reported for individuals in the Eastern South Atlantic Ocean, but again these are also larger than those in the present study, weighing up to 155 kg (Watling et al., 1981). This species is known for its extensive migrations in the Atlantic Ocean with a wide distribution and resulting in a wide range of foraging habitats, which contributes to a variable Hg bioaccumulation (Kohler et al., 2002). *I. oxyrinchus* is considered a target species for global fishing activity, which is the main pressure affecting its stocks (Rigby et al., 2019. This species is considered cosmopolitan and presents distribution in tropical and subtropical areas of the Atlantic and Pacific oceans in latitudes between 50º N - 50º S (Compagno, 2005). Depth distribution ranges from 0 to 750 m, but is usually found between 100 m and 150 m of depth (Weigmann, 2016). In Southwestern Atlantic, we only found one study reporting lower concentrations (384 ± 246 $ng.g^{-1}$) compared to our study (Mársico et al., 2007). In the Pacific ocean, one study reported Hg concentrations between 2,500 $ng.g^{-1}$ and 5,000 $ng.g^{-1}$ (Kim et al., 2016), which is higher than those in the present study, but similar to the highest Hg concentration observed for that species in the present study (5,159 $ng.g^{-1}$).

X. gladius is an oceanic species, with a global distribution in tropical and temperate waters (Amorim and Arfelli, 1984, Barret et al., 1994). This species presents vertical migrations during the day, reaching depths of up to 600 m, but at night it prefers shallow waters, lighting being a determining factor for its movements (Carey and Robison, 1981). Among the oceanographic variables, water temperature seems to have the greatest influence on the physiology and ecology of these animals and, consequently, on their spatial distribution (Palko et al., 1981), preferring temperate waters in the summer and returning to warmer waters in the fall (Collette, 1995). The Hg concentrations observed in the present study were remarkably high, possibly constituting the highest average ever recorded for this species considering data available for the Atlantic Ocean (Goyanna et al., 2023). A study conducted in the Bay of Fundy in the Gulf of Maine reported lower Hg concentrations (1245 ± 729 ng.g^{-1}) in heavier individuals (103.9 kg, range 564-1,485 kg) compared to the present study (Harding et al., 2018). In the Mediterranean Sea, a previous study also reported lower Hg concentrations (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).

The *T. alalunga* species is cosmopolitan with a distribution between 45º - 50ºN and 30º - 40°S (Collette et al., 2001; Collette and Nauen, 1983). This is a species that makes large migrations for feeding and reproductive purposes (Travassos, 1999). Oxygen levels and water temperature are the main factors shown to explain the distribution of *T. alalunga* irelative to water depth, which, in the Atlantic Ocean, can reach 600 m deep (Collette and Nauen, 1983). There is little information about Hg contamination in *T. alalunga* for the Atlantic Ocean as recently shown by Medieu et al. (2023) and Lacerda et al. (2024). Chouvelon et al. (2017) reported higher Hg values in individuals caught near the Southeastern Atlantic Ocean. These Individuals presented similar Hg concentrations to those in the present study despite being smaller in size (Teffer et al., 2014). In the Northwest Mediterranean Sea, individuals weighing between 13kg and 169 kg presented Hg values lower than the present study (Biton-Porsmoguer et al., 2022). Another study in the Gulf of Maine reported lower Hg concentration (580 ± 240 ng.g^{-1}) in heavier individuals (100 ± 31 kg) compared to our results (Schartup et al., 2019). Overall, Hg concentrations in the present study were similar of *T. alalunga* in the Northwest and Central Pacific Ocean, and in the Indian Ocean, but lower than individuals in the Mediterranean Sea (Médieu et al., 2023).

The species *T. obesus* is highly migratory, occurs at depths of up to 600m (Dagorn et al., 2000) and distributes from 50ºN - 50ºS (Miyake, 1990) in the Atlantic Ocean (Hazin and Travassos, 2007). It feeds in deep waters (400 - 500 m), consuming carnivorous demersal fish, crustaceans and squid (Mesquita et al., 2021; Silva et al., 2019; Vaske et al., 2012). This species presents 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). The Hg concentrations reported for individuals in the North Atlantic 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).

I albicans is a migratory species, which shows rapid growth and voracious carnivorous behavior even during the juvenile stage (Collette et al., 2006). Taxonomically, this species is synonymous with *I. platypterus* according to Collette et al. (2006). Its diet consists mainly of mollusks, especially cephalopods, along with fish from mesopelagic zones (Arizmendi-Rodríguez et al., 2006). Atlantic sailfish preferentially inhabit areas above the thermocline, typically found in waters ranging from 20 - 40 meters deep, with temperatures ranging from 25 - 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). We found only one study reporting Hg contamination in the Atlantic Ocean (Barber and Whaling, 1983), but none for the south region. Hg concentrations (560 ± 40 $ng.g^{-1}$) in individuals from the Eastern Pacific are similar to those of the present study for individuals with the same body size (26.4 ± 0.5 kg) (Bergés-Tiznado et al., 2015). In contrast, higher Hg values (1,480 $ng.g^{-1}$) were observed for larger individuals (49 - 168 kg) in Southeast Gulf of California (Soto-Jiménez et al., 2010). As these are different species and are patented in different locations, any deeper analysis of the data is hampered.

T. albacares is an oceanic species that 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 is not found in the Mediterranean Sea (Collette and Nauen, 1983). Mature individuals use a large part of the water column and can enjoy both the surface and deeper areas (Zavala-Camim, 1987). In general, the species prefers warmer waters compared to other tuna species, which means that their numbers increase as they get closer to the equator (Hazin, 2006). A study with *T. albacares* carried out in the Northwest Atlantic Ocean found higher Hg concentrations (304 ± 87 $ng.g^{-1}$) than those reported in this study, despite the fish being close in weight (Teffer et al., 2014). Studies carried out in areas near our catch area showed similar values, but the authors did not report the weight of the animals (Ferreira et al., 2012; Soares et al., 2009). Studies carried out in the Gulf of Mexico reported values close to ours (Cai et al., 2007; Senn et al., 2010), but with larger individuals from the same region the Hg concentrations were much higher (Kuklyte, 2012).

C. hippurus is an epipelagic and cosmopolitan species, which 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 30ºC and are geographically distributed between 47°N - 40°S, 180°W - 180°L (Palko et al., 1981). Stomach content analysis has shown that they mainly consume fish, squid and crustaceans (Massutí et al., 1998; Vaske-Jr and Lessa, 2004). *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 in a study carried out in the Southwest Atlantic Ocean, but they sampled smaller individuals (1.9 ± 1.6 kg, range 0.3 to 6.3 kg) (Moura Reis Manhães et al., 2020). However, in a study carried out in the northwest Atlantic Ocean, slightly higher concentrations (205 ± 172 ng.g-1) of Hg were found in individuals of smaller weight (4.7 ± 0.4 kg).

Stable isotopes in the species

No previous studies were found describing the $\delta^{15}N$ and $\delta^{13}C$ composition of the studied species in the Western Equatorial Atlantic. On the other hand, a few studies have described some of these species in other areas in the Atlantic, such as Central North Atlantic, Gulf of Mexico, and Southeastern Atlantic, which highlight the significance of our results. Reported values of $\delta^{15}N$ and $\delta^{13}C$ in *C. hippurus*, *T. alalunga*, *T. albacares*, *T. obesus* and *X. gladius* in the North Atlantic Ocean (Senn et al., 2010; Logan and Lutcavage, 2013) are in the same range to those observed in the present study for the respective species. Another study carried out with pelagic fishes from the northern Gulf of Mexico demonstrated similar values of $\delta^{15}N$ and $\delta^{13}C$ for *T. albacares* ($\delta^{15}N$: and $\delta^{13}C$:), but presented much lower values of $\delta^{15}N$ for *C. hippurus* ($\delta^{15}N$:) (Cai et al., 2007). It is worth mentioning that baseline values of $\delta^{15}N$ and $\delta^{13}C$ can vary spatially across multiple 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 a oceanic foraging habitat and upper trophic level position across different regions in the South and North Atlantic.

Comparing $\delta^{15}N$ and $\delta^{13}C$ values across the eight studied species, *I. oxyrinchus*, *X. gladius* and *P. glauca* presented the highest $\delta^{15}N$ values and the highest Hg and MeHg concentrations. This is due to the foraging capacity of prey higher in the food web, in addition, these animals presented higher weight values compared to the other species studied. *I. albicans* presented the greatest range of $\delta^{15}N$ values, suggesting a large variability in foraging behavior for this species. On the other hand, the species *T. albacares* and *C. hippurus* presented the lowest nitrogen values and were the animals with the lowest and less variable individual weights. These results are also consistent with the Hg concentrations, being the two species with the lowest Hg contamination levels. Regarding $\delta^{13}C$ values, the species *X. gladius, P. glauca* and *I. oxyrinchus* presented the highest ranges of values, respectively, while *T. alalunga* and *T. obesus* presented the lowest ranges. These values demonstrate the greater degree of habitat exploitation in sharks and *X. gladius* to the detriment of other species. In case of *X. gladius,* the morphological characteristics of the species make *X. gladius* an excellent swimmer, allowing it to carry out large migrations (Borges, 2001).

The niche area represents the breadth of foraging in oceanic top predators and is influenced by differences in diet and habitat use of each species, as well as food-web diversity and habitat isotopic composition. The species *T. albacares* presented the smallest isotopic niche area, suggesting a less variable foraging compared to the other species. Previous studies have shown that this species diet is composed of a very broad diet and can feed on fish, crustaceans and cephalopods, without specific preferences, which in a way is a strategy of survival in periods when environmental conditions and food availability are different (Vaske Júnior et al., 2003). Although there is no predominant preference for certain species, analyzes with stomach contents of *T. albacares* showed a preference for fish (83.4%) and shellfish (13.1%), with a small share of crustaceans (1.9%) (Vaske Júnior et al., 2003).

The species *I. oxyrinchus* and *X. gladius* presented the biggest isotopic niche area, suggesting a greater variation in foraging habitat and prey choice. These are top predators, shown to occupy the highest level of the food chain in the oceanic regions (Revill et al., 2009). They 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). *I. oxyrinchus* can also occasionally feed on other cartilaginous fish, mammals' marine life, crustaceans, turtles and seabirds (Compagno, 2001).

For *X. gladius,* authors characterize the southeastern Atlantic region as an important feeding area for swordfish due to the fact that there is a large number of cephalopods, mainly squid (Haimovici and Perez, 1990; Zavala-Camim, 1987). However, in the Northeast region, there are studies that reveal that fish were more frequent and abundant in the diet of Swordfish, probably because this equatorial region has lower cephalopod biomass (Vaske and Lessa, 2005).

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 these characteristics. 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 justify $\delta^{13}C$ 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 region (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 great 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 that occurred in the species studied, with the exception of *T. albacares* which showed little variability in $\delta^{15}N$ values.

Risk assessment and intake recommendations

As can be seen, 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 1,000 $ng.g^{-1}$ of Hg, established by Brazilian legislation. Among the bony fish species, *X. gladius, T. alalunga, I. albicans and T. obesus* exceeded the maximum limit of 300 $ng.g^{-1}$ of Hg established by the USEPA, but only Swordfish had an average concentration above the national 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).

Only the two studied species, *C. hippurus* and *T. albacares*, tuna non exceeded these levels for adults and children. Fish screening levels are very sensitive to the chosen fish consumption rate, which is why it can be well below the established 1.000 $ng.g^{-1}.kg^{-1}$ concentration limit for predador fish, established by many governmental institutions to protect human from Hg exposure through seafood consumption. The EDI and THQ results for the proposed scenarios also indicated a risk for human consuming these fish, as they were above the RfD (0.0001 0.0001 mg kg^{-1} day^{-1}) and greater than 1, respectively, for the majority of species, except *C. hippurus* and *T. albacares.* It should be emphasized that, for children consumers, these rates were much higher, indicating that possible negative effects may arise from chronic exposure to Hg at the tested consumption rates. Furthermore, negative effects on fetal neuro development may be associated with exposure of mothers, so pregnant women should avoid large predatory fish (Esposito et al., 2018).

We consider the FSL, EDI and THQ values to be very general and not very informative for consumers. For example, in Brazil, the national annual average fish consumption is relatively low, at about 5-10 kg per capita (FAO, 2020). However, in certain regions, such as amazonian riverine communities, fish consumption is much higher, approximately 150 kg per capita annually (Oliveira et al., 2010). Also, fish consumption has a strong cultural aspect in many human populations and is often consumed on a subsistence basis. Therefore, it is important to elaborate on fish consumption advisories, so consumers have a clear understanding of the consumption limits that are safe for them. To better inform consumers, we also calculated the maximum allowable fish intake (CR_{max}) based on the measured Hg level of each fish species of Hg of each fish species and report it here in a number of meals per month basis (CR_{mm})

The highest number of recommended meals were verified for Yellowfin tuna, which we consider a good choice of seafood. In fact, *T. albacares* is one of the most consumed tuna species in the world, totaling that it has global production with around 1.57 million tons produced in 2020, occupying 7th place overall (FAO, 2022). 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*, and I. albicans, 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. In Brazil, 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 Brazillian 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, and worldwide, to excessive Hg levels with the potential of causing harmful health effects.

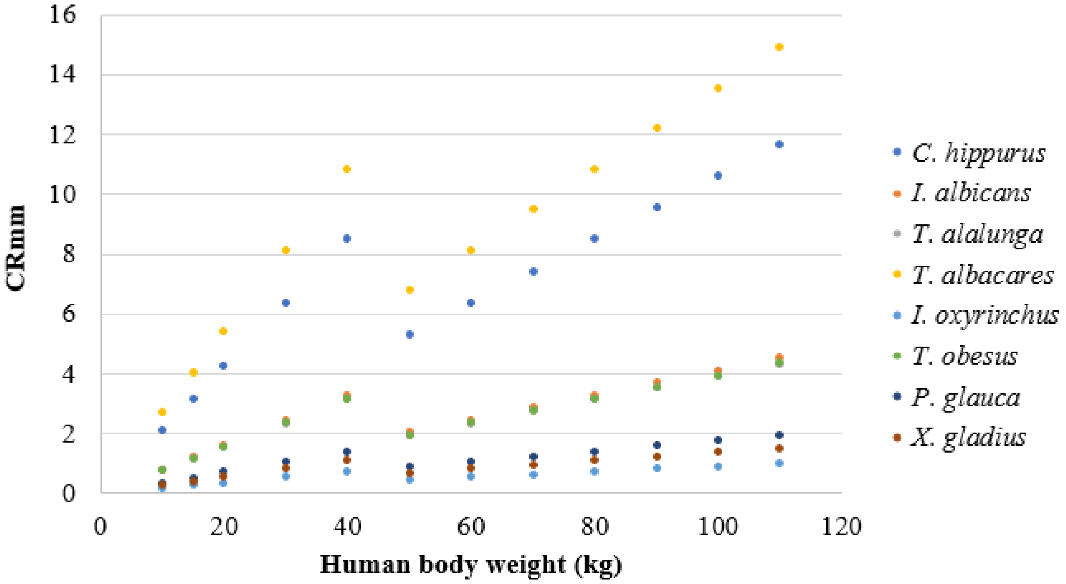

Figure 7 - Recommended number of month meals per species for children (10 to 40kg) and adults (40 to 110 kg).

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 sailffish (*I. albicans*), the Albacore (*T. alalunga*) and the Bigeye (*T. obesus*). The species the 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 shark and Swordfish species. These results show that although the eight species studied theoretically occupy the same trophic levels, there are relevant differences in different habitats, diets and foraging areas. These conditions certainly have an influence on the accumulation of contaminants, such as Hg, and partly explains the variability in accumulation between species

Finaly, it's 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 mercury (Hg). One example of the importance of this study was that children should avoid consumption of shark meat if possible. In other words, we understand that measuring the risks and benefits of different species of safe seafood consumption is mainly a question of which species are being selected for consumption and with what a question of which species are being selected for consumption and with what and how often these species are consumed.

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CONSIDERAÇÕES FINAIS E CONCLUSÕES

Em uma revisão sistêmica foi verificado as concentrações de Hg em diversas espécies de atuns e tubarões capturados no Oceano Atlântico e no Mar Mediterrâneo, na qual foram identificadas diferenças entre tubarões e peixes ósseos, com concentrações de Hg significativamente mais altas em tubarões, mas nenhuma diferença significativa foi evidenciada entre as quatro sub-regiões oceânicas avaliadas, independentemente da espécie.

A presente tese também apresentou concentrações de Hg em diversas espécies de atuns, afins e tubarões capturados no Oceano Atlântico Oeste Equatorial. Os dados gerados são inéditos para a região amostrada. Foram identificadas diferenças entre espécies, com concentrações de Hg significativamente mais elevadas nos tubarões (*I. oxyrinchus* e *P. glauca*) e no Espadarte (*X. gladius*) em comparação com Agulhão-vela (*I. albicans*), Albacora branca (*T. alalunga*), Albacora bandolim (*T. obesus*). As espécies Albacora laje (*T. albacares*) e Dourado (*C. hipurrus*) apresentaram menores concentrações entre as demais. Além disso, os resultados gerados corroboram com a literatura anterior e sugere que os peixes pelágicos oceânicos do Atlântico Sul tendem a ter maiores concentrações de mercúrio por peso e/ou tamanho em comparação com os do Atlântico Norte.

A extensa análise de isótopos estáveis (SIA) possibilitou que fossem encontradas diferenças significativas entre as espécies, que mostraram a mesma tendência observada para o Hg, ou seja, os maiores valores de 15N e 13C foram verificados para os tubarões e Espadarte. Estes resultados mostram que embora as oito espécies estudadas ocupem níveis tróficos muito próximos, existem diferenças relevantes nos diferentes habitats, dietas e áreas de alimentação que devem ser investigadas em estudos futuros. Estas condições certamente influenciam o acúmulo de contaminantes, como o Hg, e explicam em parte a variabilidade no acúmulo dentro de uma mesma espécie e entre espécies distintas.

Por último, é fundamental realçar que o consumo de peixe oferece benefícios significativos para a saúde humana, uma vez que o peixe é parte integrante de uma dieta equilibrada e é rico em ácidos gordos Ómega-3. No entanto, as recomendações fornecidas neste estudo são importantes para garantir o consumo seguro desta espécie pelos seres humanos e podem indicar uma exposição significativa ao mercúrio (Hg). Um exemplo da importância deste estudo foi que as crianças deveriam consumir tubarões com muito cuidado e evitá-los se possível. Por outras palavras, entendemos que medir os riscos e benefícios de diferentes espécies de consumo seguro de marisco é principalmente uma questão de saber quais as espécies que estão a ser selecionadas para consumo e com que frequência. essas espécies são consumidas.

APÊNDICE A – MATERIAL SUPLEMENTAR DO CAPÍTULO I

Species	Trophic level	Class	n	Hg mean (ng.g-1 w.w)	Min (ng.g-1 w.w)	Max (ng.g-1 w.w)	Above 1ppm (%)	Year of publication	Oceanic area	References
Thunnus thynnus	> 4	Osteichthyes	1292	760	250	3151	n.i.	2016	NAO	Martinez and Gay, 2016
			7	499	68	1359	n.i.	1994	MED	Pastor et al., 1994
			14	1140	n.i.	1600	n.i.	1992	NAO	Hellou et al., 1992
			169	1020	70	4260	65*/91**	2001	MED	Storelli and Marcotrigiano, 2001
			73	200	130	350	0	2005	MED	Storelli et al., 2005
			23	520	n.i.	890	0	2011	NAO	Burger and Gochfeld, 2011
			5	710	n.i.	n.i.	n.i	2019	NAO	Schartup et al., 2019
			n.i	840	n.i.	n.i.	21	2018	MED	Cammilleri et al., 2018
Thunnus albacares	> 4	Osteichthyes	52	159	48	501	0	2017	SAO	Lacerda et al., 2017
			13	327	166	531	0	2006	NAO	Besada, 2006
			7	166	93	242	0	2016	SAO	Lacerda, et al. 2016
			14	726	n.i.	n.i.	n.i.	2016	SAO	Bosch et al. 2016
			88	420	70	1200	n.i.	1973	EAO	Peterson et al., 1973
			11	61	36	90	n.i.	2006	EAO	Voegborlo et al., 2006
			8	80	3	172	n.i.	2008	SAO	Medeiros et al., 2008
			47	304	44	503	62**	2014	NAO	Teffer et al., 2014
			25	135	40	420	n.i.	2009	EAO	Costa et al. 2009
			30	n.i.	1060	1190	n.i.	1979	NAO	Greig and Krzynoweck, 1979
			56	250	68	650	n.i.	2004	NAO	Adams, 2004
			50	650	n.i.	n.i.	n.i.	2006	NAO	Burger and Gochfeld, 2006
			103	180	n.i.	n.i.	0	2007	NAO	Yan Cai et al., 2007
			18	190	n.i.	n.i.	n.i.	2010	NAO	Senn et al., 2010
			45	200	n.i.	580	11*/22**	2011	NAO	Burger and Gochfeld, 2011
			11	360	n.i.	n.i.	0	2012	NAO	Kuklyte, 2012

			56	180	10	620	0	2020	SAO	Ferreira et al., 2012
			20	120	3	280	n.i	2020	SAO	Moura Reis Manhães et al., 2020
			n.i	160	n.i.	n.i.	n.i	2018	MED	Cammilleri et al., 2018
Thunnus obesus	> 4	Osteichthyes	121	898	324	3134	36	2011	EAO	Chen et al., 2011
			30	545	95	1748	6	2017	SAO	Lacerda et al. 2017
			15	139	n.i.	n.i.	0	2016	EAO	Torres et al. 2016
			30	761	344	1290	16.7	2006	NAO	Besada, 2006
			3	231	189	274	0	2016	SAO	Lacerda, et al. 2016
			5	n.i.	230	750	0	1973	EAO	Peterson et al., 1973
			7	270	n.i.	n.i.	0	2005	NAO	Yamashita et al. 2005
Thunnus atlanticus	> 4	Osteichthyes	28	280	400	1300	n.i.	2020	SAO	Moura Reis Manhães et al. 2020
			48	640	0	1410	n.i.	2007	NAO	Cai et al. 2007
			37	1070	160	2000	n.i.	2004	NAO	Adams, 2004
			11	390	n.i.	n.i.	n.i.	2012	NAO	Kuklyte, 2012
			22	730	n.i.	n.i.	n.i.	2010	NAO	Senn et al., 2010
Thunnus alalunga	> 4	Osteichthyes	197	958	n.i.	n.i.	n.i	2017	SAO	Chouvelon et al., 2017
			15	455	294	683	87**	2014	NAO	Teffer et al., 2014
			46	370	218	1132	n.i.	1997	NAO	Andersen and Depledge, 1997
			24	190	118	564	0	2006	NAO	Besada, 2006
Katsuwonus pelamis	> 4	Osteichthyes	29	200	40	390	0	2020	SAO	Moura Reis Manhães et al., 2020
			n.i.	n.i.	80	220	n.i.	1981	NAO	Farrant et al., 1981
			53	192	89	336	n.i.	1997	NAO	Andersen and Depledge 1997
			25	115	77	144	n.i.	2017	EAO	Vieira etal., 2017
			9	109	84	219	0	2019	SAO	Mirlean etal., 2019
			132	340	150	n.i.	n.i.	1993	NAO	Armas etal., 1993
			12	152	n.i.	n.i.	n.i.	1979	NAO	Greig and Krzynoweck, 1979
			15	40	n.i.	n.i.	0	2016	EAO	Torres et al., 2016
Prionace glauca	> 4	Elasmobranchii	40	520	140	1710	10	2018	NAO	Biton-Porsmoguer et al., 2018
			27	n.i.	680	2500	n.i.	2007	EAO	Branco et al., 2007

			37	n.i.	220	1300	n.i.	2007	NAO	Branco et al., 2007
			15	2250	n.i.	n.i.	100	2015	NAO	Matos J. et al., 2015
			20	1360	n.i.	n.i.	n.i.	2013	SAO	Velez et al., 2013
			4	1900	n.i.	n.i.	n.i.	2007	SAO	Augelli et al., 2007
			8	1270	n.i.	n.i.	n.i.	2021	NAO	Hauser-Davis R. et al., 2021
			47	760	150	2300	21	2008	SAO	Dias et al., 2008
			27	1120	460	2400	40	2014	SAO	De Carvalho et al., 2014
			30	398	12	11500	n.i.	2007	SAO	Mársico et al., 2007
			5	726	280	1170	40	1999	SAO	Morales-Aizpurúa et al., 1999
			15	n.i.	210	1500	27	1995	SAO	Chicourel et al., 1995
			30	330	140	500	0	2017	EAO	Torres P. et al., 2017
			26	n.i.	160	1840	n.i.	2004	NAO	Branco et al., 2004
			23	n.i.	160	1200	n.i.	2004	NAO	Branco et al., 2004
Coryphaena hippurus	> 4	Osteichthyes	57	70	10	490	0	2007	NAO	Cai et al., 2007
			39	205	21	648	28**	2014	NAO	Teffer et al., 2014
			385	100	12	550	0	2009	NAO	Adams, 2009
			27	210	n.i.	n.i.	0	2012	NAO	Kuklyte, 2012
			27	170	710	710	n.i.	2011	NAO	Burger and Gochfeld, 2011
			20	53	26	90	0	2002	SAO	Selanes et al. 2002
			22	40	8	100	0	2020	SAO	Moura Reis Manhães et al., 2020
Xiphias gladius	> 4	Osteichthyes	192	620	40	2210	14	2001	SAO	Mendez at al., 2001
			162	490	150	1050	4.3	2001	MED	Storelli and Marcotrigiano, 2001
			58	70	20	150	0	2005	MED	Storelli et al., 2005
			7	470	n.i.	820	0	2005	n.i.	Yamashita et al. 2005
			9	67	65	70	0	2019	SAO	Mirlean etal., 2019
			n.i.	1340	n.i.	n.i.	n.i.	2019	SAO	Buck et al., 2019
			27	958	410	2110	37	2010	n.i.	Escribano S. and Montoro R., 2010
			107	630	n.i.	n.i.	n.i.	2012	SAO	Rodrigues M. et al., 2012
			585	550	n.i.	n.i.	n.i.	2012	SAO	Rodrigues M. et al., 2012

			23	n.i.	900	2200	n.i.	2007	EAO	Branco et al., 2007
			29	n.i.	31	2400	n.i.	2007	NAO	Branco et al., 2007
			17	816	120	2000	29.4	2018	SAO	M. Esposito et al., 2018
			17	672	164	1350	17.7	2018	EAO	M. Esposito et al., 2018
			11	667	53	1520	18.2	2018	SAO	M. Esposito et al., 2018
			25	475	33	2140	8	2018	NAO	M. Esposito et al., 2018
			20	615	120	1660	5	2018	MED	M. Esposito et al., 2018
			26	660	n.i.	n.i.	n.i.	2022	NAO	Biton-Porsmoguer et al., 2022
			11	887	n.i.	n.i.	n.i.	2011	NAO	Damiano S. et al., 2011
			11	658	n.i.	n.i.	n.i.	2011	NAO	Damiano S. et al., 2011
			136	1067	60	4910	71*	1990	NAO	Monteiro and Lopes, 1990
			30	473	n.i.	n.i.	n.i.	2015	EAO	Zaza S. et al., 2015
			18	1400	150	3070	67	2006	NAO	Burger and Gochfeld, 2006
			48	620	200	1300	10.4	2008	SAO	Dias et al., 2008
			11	580	n.i.	n.i.	n.i.	2019	NAO	Schartup et al., 2019
			3	417	340	470	n.i.	1994	MED	Pastor et al., 1994
Isurus oxyrinchus	> 4	Elasmobranchii	4	840	350	1530	1	2017	EAO	Torres P. et al., 2017
			9	502	472	530	0	2019	SAO	Mirlean et al., 2019
			51	1830	n.i.	6210	80/86*/88**	2011	SAO	Burger and Gochfeld, 2011
			32	2647	755	4933	100**	2014	SAO	Teffer et al., 2014
			19	2110	590	5580	79	1981	SAO	Watling and Stanton, 1981
			1	510	n.i.	n.i.	n.i.	1999	SAO	Morales-Aizpurúa et al., 1999
			4	384	120	691	n.i.	2007	SAO	Mársico et al., 2007
			48	740	120	2570	25	2018	NAO	Biton-Porsmoguer et al., 2018
Euthynnus alletteratus	> 4	Osteichthyes	8	210	20	380	n.i.	2020	SAO	Moura Reis Manhães et al., 2020
			9	690	n.i.	n.i.	n.i.	2012	NAO	Kuklyte, 2012
			9	1080	240	2520	n.i.	2007	NAO	Cai et al. 2007
			114	1070	160	2000	18/75*	2004	NAO	Adams, 2004

Legends: *percentage of values above 0.5ppm **percentage of values above 0.3ppm

NAO	North Atlantic Ocean
SAO	South Atlantic Ocean
EAO	Equatorial Atlantic Ocean
MED	Mediterranean Sea

APÊNDICE B – MATERIAL SUPLEMENTAR DO CAPÍTULO II

Table S.1. Fish weight and estimated fork length (SFL), stable isotope composition ($\delta^{13}C$ and $\delta^{15}N$) and total Hg concentrations of *Thunnus alalunga* sampled the Western Equatorial Atlantic Ocean. Date: * August 2020; all others from October 2022.

Sample	Total weight (kg)	Estimated SFL (cm)	Total Hg (ng g^{-1} w.w.)	$\delta^{13}C$	$\delta^{15}N$
1	28.6*	62.9	677	--	--
2	26.65*	62.4	482	-17.18	10.02
3	28.6*	62.9	610	-17.36	10.61
4	29.9*	63.3	704	-17.23	11.43
5	28.73	55.4	386	-17.39	10.77
6	23.01	61.4	469	-17.60	11.20
7	26.91	62.5	377	-17.35	9.80
8	22.1	61.1	362	-17.38	11.78
9	26	62.2	378	-17.78	11.76
10	23.4	61.5	430	--	--
11	31.07	63.6	512	-17.76	9.80
12	29.77	63.3	554	-17.24	11.60
13	25.61	62.1	648	-17.11	12.28
14	28.86	63.0	605	-17.36	10.69
15	34.71	64.6	572	-17.79	11.42
16	25.09	62.0	431	-17.36	11.20
17	44.59	67.4	930	-17.98	10.65
18	35.49	64.9	655	-17.32	10.70
19	28.34	62.9	639	-17.51	11.07
20	26	62.2	454	-17.57	12.76
21	32.5	64.0	759	-17.25	10.96
22	32.89	64.1	627	-17.47	11.21
23	27.82	62.7	496	-17.03	11.02
24	34.58	64.6	753	-17.38	10.12
25	32.5	64.0	657	-17.62	12.01
26	21.58	61.0	312	-17.94	12.06
27	27.04	62.5	514	-17.31	12.18
28	20.8	60.8	345	-17.87	11.89
29	33.54	64.3	632	-17.06	11.44
30	27.82	62.7	460	-17.03	11.02
31	36.27	65.1	779	-17.00	11.19
32	27.95	62.8	443	-17.44	11.92
33	26.26	62.3	508	-17.70	10.82
34	28.08	62.8	413	-17.68	11.30
35	26.52	62.4	404	-17.34	11.30
36	24.31	61.8	443	-17.82	11.30
37	32.5	64.0	497	-17.34	10.71
38	28.08	62.8	318	-17.76	11.81
39	25.87	62.2	447	-17.41	11.26

40	23.79	61.6	438	-17.82	11.33
41	24.05	61.7	340	-17.44	13.28
42	30.29	63.4	552	-17.02	10.08
43	27.3	62.6	294	-17.12	11.26
44	29.12	63.1	342	-17.82	12.35