Marine Pollution Bulletin 203 (2024) 116469

Contents lists available at ScienceDirect

Marine Pollution Bulletin

journal homepage: www.elsevier.com/locate/marpolbul

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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https://doi.org/10.1016/j.marpolbul.2024.116469
Received 7 December 2023; Received in revised form 2 May 2024; Accepted 3 May 2024
Available online 15 May 2024

the four most exploited tuna species worldwide and is one of the main 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 was 52,519 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 and 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 ($^{13}C/^{12}C$, hereafter $\delta^{13}C$) and nitrogen ($^{15}N/^{14}N$, hereafter $\delta^{15}N$) 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 $\delta^{13}C$ (~1 ‰) and $\delta^{15}N$ (~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 16°- 30° W (Fig. 1). These catches are mostly constituted by *Thunnus albacares* (34 %) and *Xiphias gladius* (31 %), but *T. alalunga* represents about 4 % of catches 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).

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

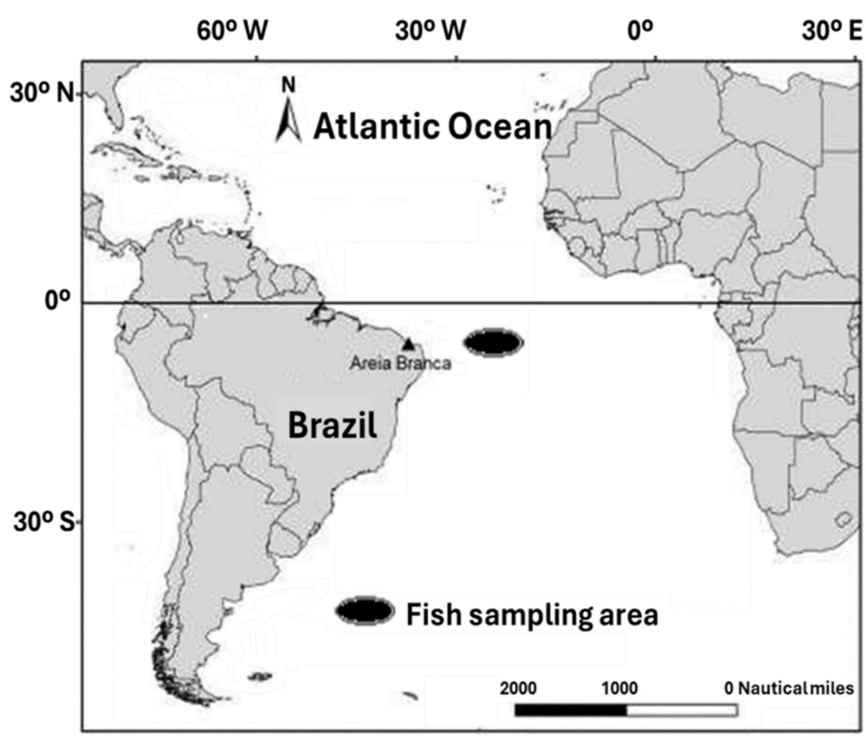

Fig. 1. Map showing the approximate distribution of fishing areas from where *T. alalunga* was obtained for this study.

plastic zip lock bags, frozen at a minimum of −20 °C, 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 (HNO_3, 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 (H_2O_2, 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 ($SnCl_2$) prepared in 10 % sulfuric acid (20 % H_2SO_4). 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 S1.

Subsamples 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 °C 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 Developemt Core Supply Team, 2021). Data normality was tested using the Shapiro-Wilk test. All tests were conducted assuming a significance level of 99 % ($p < 0.05$).

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 S1).

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 and 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 and Depledge, 1997).

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 to −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

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).

Stable isotope composition ($\delta^{13}C$: −17.0 ‰ to −18.0 ‰ and $\delta^{15}N$: 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 (Dulvy 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 (Dulvy 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 (Table 1) also reflects a high trophic position and is similar to *T. albacares* (Voegborlo et al., 2006) and *T. obesus* (Médieu et al., 2022,

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 and 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)

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; $r^2 = 0.596$; $p < 0.05$; $n = 44$) (Fig. 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 – 3074; $r^2 = 0.445$; $p < 0.05$; $n = 44$.

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).

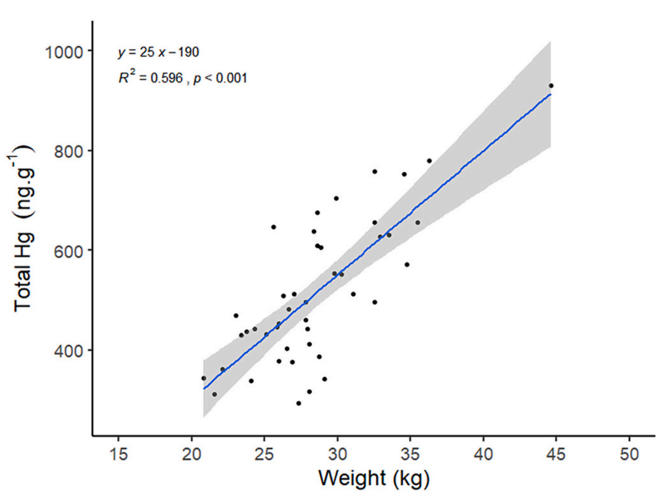

Fig. 2. Relationship between total Hg concentrations and fish weight in *T. alalunga* caught in the Equatorial Southwestern Atlantic.

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_{bw}^{-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 and 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 Wester 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.

Supplementary data to this article can be found online at https://doi.org/10.1016/j.marpolbul.2024.116469.

CRediT authorship contribution statement

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

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper. All data used in this paper is available as supplementary material.

Data availability

All data used are available as supplementary material

Acknowledgements

The authors would like to thank members of the Coastal Biogeochemistry Laboratory (UFC/LABOMAR) for helping with the sampling and analysis. This research was funded by Fundação Cearense de Apoio ao Desenvolvimento Científico e Tecnológico (FUNCAP) Project No. UNI-0210-00030.01.00/23 and Grant. No. 11276248/2021. This study is an output of the INCT Continent-Ocean Materials Transfer (INCT-TMC Ocean partially supported by Conselho Nacional de Desenvolvimento Científico e Tecnológico - CNPq Proc. No. 405.765/2022–3, and Proc. No. 573.601/2008–2 and 304.140/2023-6 to LD Lacerda. CE Rezende received financial support from CNPq and FAPERJ.

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