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Species-specific mercury speciation in billfishes and its implications for food safety monitoring and dietary advice

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ABSTRACT

Humans are exposed to toxic methylmercury mainly by consuming marine fish, in particular top predator species like billfishes or tunas. In seafood risk assessments, mercury is assumed to be mostly present as organic methylmercury in predatory fishes; yet high percentages of inorganic mercury were recently reported in marlins, suggesting markedly different methylmercury metabolism across species. We quantified total mercury and methylmercury concentrations in muscle of four billfish species from the Indian and the Pacific oceans to address this knowledge gap. We found low percentages of methylmercury in blue and black marlins (15 ± 7 %) compared to swordfish and striped marlin (89 ± 13 %), with no significant differences among ocean regions. This illustrates that billfishes exhibit species-specific methylmercury bioaccumulation patterns, likely related to unique selenium-dependent *in vivo* methylmercury demethylation capacities in muscle. Blue and black marlins therefore appeared generally safer for human consumption than swordfish and striped marlin regarding MeHg toxicological effects. Yet, no matter the species, the frequency of recommended weekly billfish meals decreased with increasing fish size, given that mercury naturally accumulates over time. When assessing potential risks of billfish consumption, we therefore recommend measuring methylmercury, rather than total mercury, and relying on a large number of samples to cover a broad range of fish sizes. This study calls for additional characterization of mercury speciation and bioavailability in billfishes to better understand the mechanisms driving species-specific differences of methylmercury detoxification, and to refine dietary advices associated to marine top predators consumption.

1. Introduction

Mercury (Hg) is a widespread heavy metal of particular concern to wildlife and human health, especially its organic form, namely methylmercury (MeHg). In oceans, inorganic Hg can be microbially converted into MeHg, which is partly incorporated into primary producers and biomagnified into marine food webs, leading to elevated MeHg proportions (%MeHg) and concentrations in higher-trophic-level organisms. Humans are exposed to MeHg mainly by their consumption of seafood (Sunderland et al., 2009), in particular predatory fishes like tunas and billfishes (i.e., pelagic fishes with a long, bony, and spear-shaped bill, of families Xiphiidae and Istiophoridae), in which MeHg generally represent the major chemical form (> 90 %) of total Hg in muscle. Yet, high proportions of inorganic Hg over MeHg were reported in blue marlin (*Makaira nigricans*) (Manceau et al., 2021a; Shultz and Crear, 1976), a billfish commonly consumed in tropical regions,

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suggesting different demethylation capabilities across top predatory fishes. With the adoption of the United Nations Environment Program Minamata Convention and its objective of protecting human health from Hg harmful effects, it has become a priority to document Hg content in marine commercial species, and to assess MeHg exposure to humans through seafood consumption.

In human populations, chronic exposure to MeHg from diet during pregnancy have been associated to negative neurodevelopmental outcomes at birth and in children, while exposure later in life can lead to neurological system impairments, with evidence of cardiovascular and immune system impacts as well (Basu et al., 2023; Ha et al., 2017). To limit risks on human health of MeHg exposure, consumption guideline levels for MeHg were set at 1.0 $\mu g.g^{-1}$ wet weight (ww) for predatory fishes such as billfishes and tunas, and 0.5 $\mu g.g^{-1}$ ww for other fish species (FAO and WHO, 2005). As Hg toxicological risk not only relies on concentrations in fishes, but also on the frequency of consumption and on the weight of the consumer, provisional tolerable weekly intake (PTWI) were also developed, corresponding to the estimated amount per unit of body weight (bw) of Hg that can be ingested over a week without risk of adverse health effects. For fish and shellfish products, the PTWI for MeHg was first set at 3.3 μg/kg bw by the Joint FAO/WHO Expert Committee on Food Additives (JECFA), before being revised to 1.6 μg/kg bw, based on epidemiological studies investigating the link between MeHg exposure in mothers and the subsequent neurological development effects in cohorts of children from the Faroe Islands and the Seychelles (FAO and WHO, 2007). A re-evaluation of these cohorts by the European Food Safety Authority even suggested a revised PTWI at 1.3 μg of MeHg/kg bw (EFSA, 2012).

While guideline levels and PTWI values were defined regarding fish MeHg content, potential risk assessments associated to pelagic predator consumption are generally based on total Hg concentrations, assuming that most of Hg is usually in its organic form in muscle tissues of predatory fishes (Bille et al., 2020; Soto-Jiménez et al., 2010). Compared to MeHg analysis, the analysis of total Hg concentrations is more cost and time effective, and accessible to those without advanced and expensive laboratory facilities. It therefore offers the possibilities to evaluate Hg exposure in many world's biomes and ecosystems, as requested by the Minamata Convention (AMAP, Un Environment, 2019). Yet, this general assumption is now challenged by low %MeHg reported in blue marlin in Hawaii (15 ± 10 %; Shultz and Crear, 1976) and French Polynesia (19 ± 22 %; Manceau et al., 2021a). These low %MeHg have been suggested to result from *in vivo* MeHg demethylation in blue marlin muscle tissue itself, through the biomineralization of MeHg-cysteine complexes (MeHgCys) into inorganic Hg-selenium (Se) complexes, mostly as Hg-tetraselenolate [$Hg(Sec)_4$] and Hg selenite (HgSe) (Manceau et al., 2021a). This mechanism has also been observed in other tissues and marine organisms such as the liver and brain of pilot whale (Gajdosechova et al., 2016) and the liver, kidneys, muscle, and brain of giant petrel seabirds (Manceau et al., 2021b). Contrary to MeHg forms that are highly bioavailable (i.e., easily absorbed by the gastrointestinal tract) for humans (Bradley et al., 2017), Hg-Se complexes are suspected to be relatively stable and either not bioavailable, or exhibiting little to no toxicity if bioavailable (Takahashi et al., 2021), although further research remains needed to better characterize the bioavailability of all Hg-Se complexes. Using total Hg concentrations instead of MeHg concentrations therefore overestimates human exposure to MeHg associated to blue marlin consumption and might lead to more restrictive dietary advices than necessary. Such biased dietary advices could be detrimental to the health and wellbeing of food insecure coastal populations where local fish consumption has been declining at the expense of a Western-style diet, while remaining highly recommended to fight non-communicable diseases (e.g., diabetes) and obesity (Bell et al., 2015; Wesolowska et al., 2024).

As pelagic fishes, billfish species display relatively high levels of proteins, omega-3 fatty acids, vitamins, and minerals, and represent a potential major source of essential nutrients for tropical coastal communities (Bodin et al., 2017; Sardenne et al., 2020; Sioen et al., 2009; Sirot et al., 2012; Wesolowska et al., 2024). Billfishes are mainly caught by industrial fisheries (using longlines and gillnets, and as bycatch of purse seine) in offshore areas (Peatman et al., 2023), but also by recreational and artisanal gears in more coastal fishing grounds (IOTC Secretariat, 2022), and have therefore a high economic and cultural value in tropical areas where they are distributed (Kadagi et al., 2022). In this context, quantifying MeHg concentrations and %MeHg in billfishes appears essential to determine more precisely to what extent these species can be safely consumed, and to provide appropriate dietary advices.

We measured total Hg and MeHg concentrations, alongside total Se concentrations, in four different billfish species to i) explore the extent of species-specific *in vivo* demethylation capabilities, and ii) revisit the implication for potential risk to human health resulting from these billfishes consumption. Our study focuses on white dorsal muscle tissue as it is the most frequently consumed part of predatory fishes, and on four billfish species that are harvested and consumed in both the Indian and the Pacific Oceans: swordfish (*Xiphias gladius*), striped marlin (*Kajikia audax*), blue marlin (*Makaira nigricans*), and black marlin (*M. indica*). We hypothesised that some billfish species – particularly blue marlin – are capable of *in vivo* demethylation, likely involving a Se-mediated pathway, and resulting in lower risk to human health associated with their consumption.

2. Material and methods

2.1. Billfish sample collection

A total of 45 billfish samples were collected onboard pelagic semi-industrial longliners and recreational trolling boats in five regions of the western Indian and the southwestern Pacific (Fig. 1). In the Indian Ocean, it corresponds to eight swordfish collected in the Seychelles archipelago (in 2013 and 2014), four swordfish around Reunion Island (2013–2014), eight swordfish off South Africa (2015), and nine blue marlins around the island of Mauritius (2021–2022). In the Pacific, one swordfish, five striped marlins, seven blue marlins, and three black marlins, were collected in the vicinity of New Caledonia in 2021 (Table 1). All individuals were measured to the lowest cm (lower jaw fork length, i.e., from the tip of the lower jaw to the fork of the tail), with individual sizes per species and ocean region reported in Table 1. For all billfishes, a white muscle sample of around 10 g ww was collected in the front dorsal position, stored frozen at −20 °C, freeze-dried for 72 h, and ground to a fine homogeneous powder.

2.2. Laboratory analyses

2.2.1. Total mercury and methylmercury concentrations

Homogenized freeze-dried billfish muscle samples were analysed for total Hg and MeHg at GET (Toulouse, France). Total Hg concentrations were measured on triplicates of 2–10 mg aliquots by thermal decomposition, gold amalgamation, and atomic absorption (DMA-80, Milestone, Italy). Blanks and the certified reference material (CRM) BCR-464 (tuna fish muscle; total Hg = 5.24 ± 0.10 $\mu g.g^{-1}$ dry weight, dw) were routinely used every 5–10 samples, displaying an average recovery of 100 ± 3 % (n = 35) among the different analytical batches. The precision of total Hg measurements was below 9 % for n = 3 replicates of each sample.

To determine MeHg concentrations, 100 ± 10 mg of muscle samples were digested with 5 mL of 6 N aqueous solution of nitric acid (trace metal grade, HNO_3) at 85 °C on a hot plate during 8 h. Sample aliquots were diluted in Milli-Q water to approximately 0.1 $ng.g^{-1}$, and buffered to pH 4.9 ± 0.1 with the addition of 0.3 mL of 2 M acetate buffer. Fifty μL of a 1 % sodium tetraethylborate ($NaBEt_4$) solution were added to all samples before purge and trap gas chromatography atomic fluorescence spectrometry analysis (MERX[TM], Brooks Rand), following the EPA

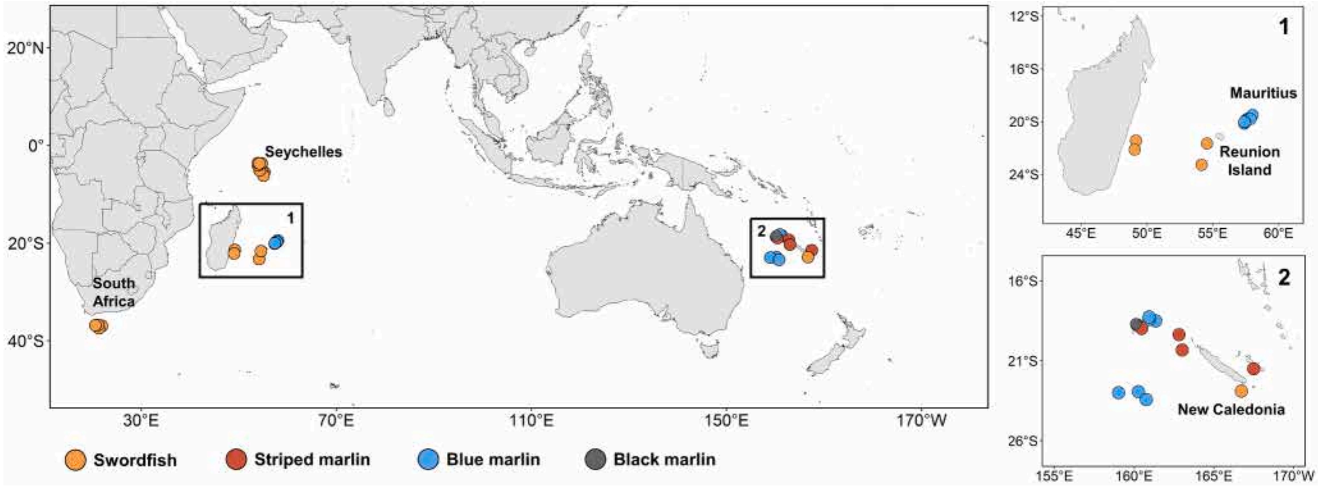

Fig. 1. Sample location of swordfish (orange), striped marlin (red), blue marlin (blue), and black marlin (dark grey) in the western Indian and southwestern Pacific oceans. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

Table 1
Characteristics of billfish samples, concentrations of total mercury, methylmercury and total selenium. Mean ± standard deviation (minimum – maximum) of fish size (lower jaw fork length, cm), water content (%), total lipid content (TLC, %), total mercury (THg), methylmercury (MeHg), and total selenium (TSe) concentrations (μg.g^{-1}, ww), and percentage of methylmercury (%MeHg, %) in billfish muscle samples. n gives the number of analysed individuals.

Species	Ocean	Ocean region	*n*	Size (cm)	Water content (%)	TLC (%)	THg (μg.g^{-1}, ww)	MeHg (μg.g^{-1}, ww)	%MeHg (%)	TSe (μg.g^{-1}, ww)
Swordfish	Indian	Seychelles	8	169 ± 42 (110 – 230)	–	24 ± 17 (3 – 52)	1.03 ± 0.58 (0.21 – 1.79)	0.88 ± 0.43 (0.23 – 1.38)	90 ± 10 (77 – 109)	0.82 ± 0.39 (0.46 – 1.69)
		Reunion Islands	4	120 ± 39 (87 – 171)	–	21 ± 19 (4 – 40)	0.47 ± 0.38 (0.15 – 0.99)	0.40 ± 0.29 (0.16 – 0.81)	91 ± 13 (79 – 107)	0.50 ± 0.03 (0.46 – 0.53)
		South Africa	8	168 ± 35 (131 – 230)	–	43 ± 17 (19 – 70)	0.70 ± 0.26 (0.27 – 1.13)	0.58 ± 0.22 (0.32 – 0.90)	85 ± 19 (67 – 117)	0.59 ± 0.21 (0.26 – 0.87)
	Pacific	New Caledonia	1	102	74	6	0.56	0.58	103	0.89
Striped marlin	Pacific	New Caledonia	5	218 ± 17 (196 – 247)	75 ± 3 (71 – 81)	12 ± 5 (8 – 18)	0.71 ± 0.45 (0.35 – 1.17)	0.64 ± 0.42 (0.31 – 1.07)	90 ± 8 (81 – 103)	0.77 ± 0.10 (0.67 – 0.93)
Blue marlin	Indian	Mauritius	9	217 ± 41 (170 – 288)	75 ± 1 (72 – 77)	6 ± 3 (3 – 11)	2.32 ± 2.07 (0.62 – 7.49)	0.31 ± 0.23 (0.09 – 0.80)	15 ± 8 (7 – 34)	–
	Pacific	New Caledonia	7	246 ± 53 (189 – 315)	75 ± 3 (69 – 77)	8 ± 5 (5 – 18)	4.58 ± 2.90 (1.30 – 9.23)	0.42 ± 0.29 (0.07 – 0.94)	9 ± 5 (5—–19)	2.31 ± 1.08 (1.16 – 3.99)
Black marlin	Pacific	New Caledonia	3	242 ± 20 (219 – 257)	76 ± 1 (75 – 77)	11 ± 10 (4 – 22)	4.49 ± 1.12 (3.23 – 5.38)	0.67 ± 0.32 (0.44 – 1.03)	15 ± 5 (9 – 19)	2.22 ± 0.37 (1.80 – 2.49)

Method 1630 (U.S. Environmental Protection Agency, 1998). Muscle tissues were analysed in triplicate, with analytical blanks and diluted CRM standard solution included every three samples to check MeHg measurement accuracy. The CRM BCR-464 (MeHg = 5.50 ± 0.17 μg.g^{-1} dw) used showed an average recovery of 96 ± 11 % (n = 16) among the different analytical batches.

All total Hg and MeHg concentrations obtained on freeze-dried materials were converted on a ww basis, using a mean water content of 75 % (Table 1), to be compared to safety guidelines. Percentage of MeHg (%MeHg) was defined as the ratio between MeHg and total Hg concentrations, and percentage of inorganic Hg (%iHg) was the difference between 100 % and %MeHg. Molar concentrations of total Hg and MeHg were calculated dividing the concentrations (μg.g^{-1}) of total Hg and MeHg by the atomic mass of Hg (200.59) and MeHg (215.63), respectively. Molar concentrations of inorganic Hg were the differences between molar concentrations of total Hg and MeHg.

2.2.2. Total selenium concentrations

Except for blue marlins from Mauritius, homogenized freeze-dried billfish muscle samples were analysed for total Se in different study-specific laboratories with analytical blanks and laboratory-specific reference standards. For swordfish from the western Indian Ocean, aliquots of 50–300 mg were digested with a mixture of hydrochloric and nitric acids in a microwave and total Se concentrations were analysed by inductively coupled plasma atomic emission spectrometry on a Varian Vista-Pro ICP-OES at the LIENSs facility (La Rochelle, France) (Bodin et al., 2017). The two CRMs TORT-3 (lobster hepatopancreas, total Se = 10.9 ± 1.0 μg.g^{-1} dw) and DOLT-4 (dogfish liver, total Se = 8.3 ± 1.3 μg.g^{-1} dw) (n = 17) showed an average recovery of 103 ± 1 % and 102 ± 1 %, respectively. For billfish samples from the western Pacific Ocean, 30 ± 5 mg aliquots were digested at 80 °C for 3 h in closed 30-mL Teflon screw-cap vials (Savillex, Minnetonka, MN, USA) with 1 mL 65 % nitric acid (Suprapur, Merck) and 0.25 mL 30 % hydrogen peroxide (Suprapur, Merck), before being diluted ten times in Milli-Q water and transferred into 15-mL polypropylene pre-washed tubes. Total Se concentrations were determined at LEMAR and PSO (Plouzané, France) on a sector field mass spectrometer (SF-ICP-MS, Thermo Fisher Scientific Element XR) using the signal obtained at high resolution for the ^{77}Se isotope. The two CRMs TORT-3 and NIST-2976 (mussel tissue, total Se = 1.73 ± 0.19 μg.g^{-1} dw) showed an average recovery of 95 ± 2 % (n = 5) and 96 ± 11 % (n = 5), respectively.

As for total Hg and MeHg levels, all total Se concentrations were converted on a ww basis considering a mean water content of 75 %. Molar concentrations of total Se were calculated dividing these concentrations (μg.g^{-1}) by the atomic mass of Se (78.96).

2.2.3. Total lipid content

We quantified the total lipid content to investigate its influence on billfish Hg levels. For each billfish sample, 150 ± 10 mg of a homogenized dried aliquot were extracted twice with 20 mL of solvent (dichloromethane and methanol (2:1)) at 100 °C under 1400 psi for 5 min using a ASE 350 Accelerated Solvent Extractor (Dionex, Voisins de Bretonneux, France) at LEMAR (Plouzané, France), following the methods of Bodin et al. (2009). Lipid extracts were evaporated to dryness under N_2 flow with a N-evap 111 nitrogen evaporator (OA-SYS, Berlin, USA) to determine the total lipid content expressed in % of dry weight tissue.

2.3. Statistical analyses

Data analyses were performed with the statistical open source R software 3.6.1 (R Core Team, 2018). Total Hg and MeHg concentrations were first log-transformed to guarantee the homogeneity of variance in statistical analyses (Zuur et al., 2010). Analysis of Covariance (ANCOVA) was used to investigate the effect of species, ocean region, individual fish size, and individual lipid content on log-transformed total Hg and MeHg concentrations, and on %MeHg in muscle tissues.

For swordfish and blue marlin (i.e., species with more than 10 samples), we calculated species-specific bioaccumulative curves to illustrate the natural bioaccumulative processes of Hg in individuals through time. This consisted in fitting species-specific power relationships ($\log(\mathrm{Hg}) = \mathrm{a} \times (\mathrm{LJFL} - \mathrm{c})^{b} - \mathrm{d}$) between log-transformed total Hg or MeHg concentrations and fish size (LJFL) (Houssard et al., 2019; Médieu et al., 2022, Médieu et al., 2021), combining data from all ocean regions as no regional differences were found in our data (Kruskal-Wallis, $p > 0.05$ for the two species).

Finally, we fitted species-specific linear regressions between inorganic Hg and total Se molar concentrations to investigate species-specific Hg-Se patterns in relation to different MeHg metabolism.

2.4. Toxicological risk assessment

For all billfish individuals, we calculated the quantity of fish (Q, g) at which PTWI is reached, i.e., the amount of fish above which consumption may pose health problems regarding MeHg intake. For this calculation, we assumed that i) MeHg is highly bioavailable (Bradley et al., 2017), and that ii) in species with high proportions of inorganic Hg over MeHg, inorganic Hg is mainly complexed as inert and poorly bioavailable HgSe nanoparticles, as recently evidenced in blue marlin muscle (Manceau et al., 2021a; Takahashi et al., 2021). We therefore calculated the quantity of fish Q at which PTWI is reached as follow:

$$Q = PTWI_{MeHg} / [MeHg] \times BW$$

where [MeHg] are the individual fish muscle MeHg concentrations ($\mu g.g^{-1}$, ww), and BW the consumer body weights, set at 30 ± 20 kg for children, and 75 ± 25 kg for adults (including pregnant and/or breastfeeding women). $PTWI_{MeHg}$ is the PTWI value for MeHg, arbitrarily set at 1.3 µg/kg bw, i.e., the most restrictive PTWI value based on the latest evaluation of the children cohorts in Seychelles and the Faroe Islands (EFSA, 2012).

We then compared these amounts of fish Q to a mean fish portion to estimate the maximum recommended number of fish meals per week. We considered mean fish portions of 150 g and 75 g for adults and children, respectively, as recommended by the Government of New Caledonia (ANSES, 2019).

3. Results and discussion

3.1. Total mercury and methylmercury concentrations in billfish muscle

Total Hg concentrations were highly variable among species and ocean regions, ranging from 0.15 $\mu g.g^{-1}$ ww in swordfish from Reunion Island to 9.23 $\mu g.g^{-1}$ ww in blue marlin from New Caledonia (Fig. 2, Table 1). Highest total Hg concentrations were measured in blue and black marlins from New Caledonia, with mean values four to nine times higher than the ones measured in swordfish and striped marlin (all ocean regions combined). Blue marlin caught around Mauritius also displayed high total Hg concentrations, with mean values two to five times higher than in swordfish and striped marlin. These highly variable and sometimes elevated Hg concentrations measured in all species are in accordance with what is reported in the literature for billfishes from different regions of the global ocean (SI Appendix Table S1).

Most of the total Hg content variability was explained by fish size differences (Fig. 3A; ANCOVA, $p < 0.05$ for fish size only), with highest total Hg concentrations in blue and black marlins associated to larger individuals. This natural bioaccumulative property with age is well

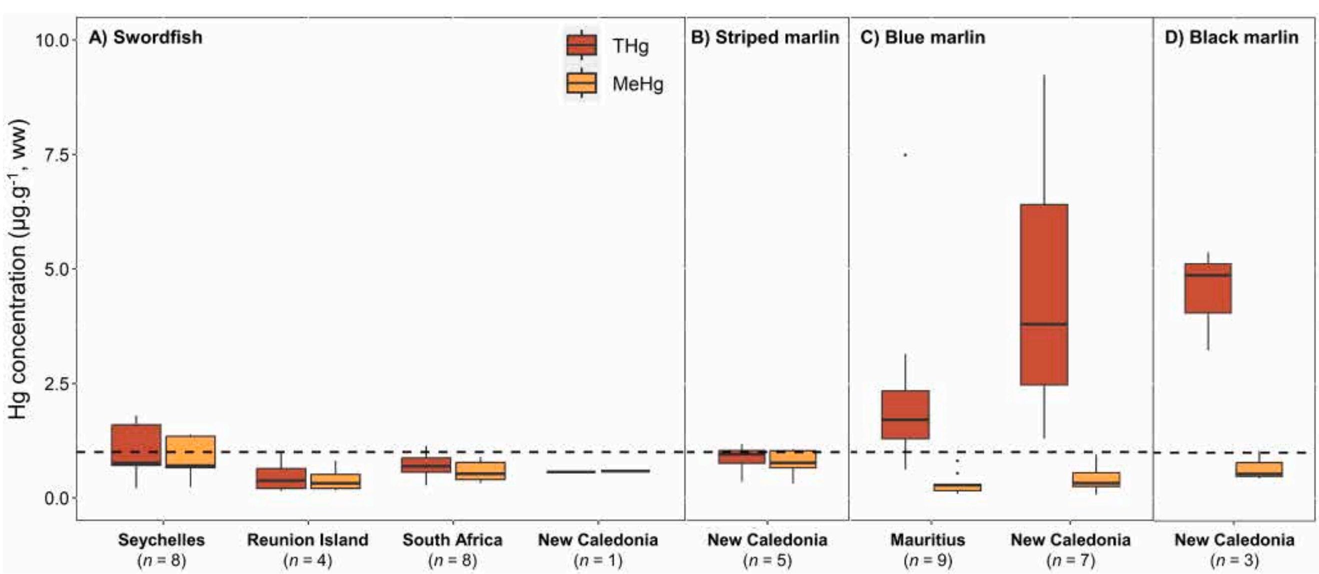

Fig. 2. Variability of mercury content in billfish muscle. Boxplots of total mercury (THg) and methylmercury (MeHg) concentrations ($\mu g.g^{-1}$, ww) in muscle samples of **A)** swordfish, **B)** striped marlin, **C)** blue marlin, and **D)** black marlin, caught in different regions of the Indian (i.e., Seychelles, Reunion Island, South Africa, and Mauritius) and the Pacific (i.e., New Caledonia). The dashed line represents the MeHg consumption guideline value for large predatory fish of 1 $\mu g.g^{-1}$ ww (FAO and WHO, 2005)(FAO Codex Alimentarius Commission, 2019). (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

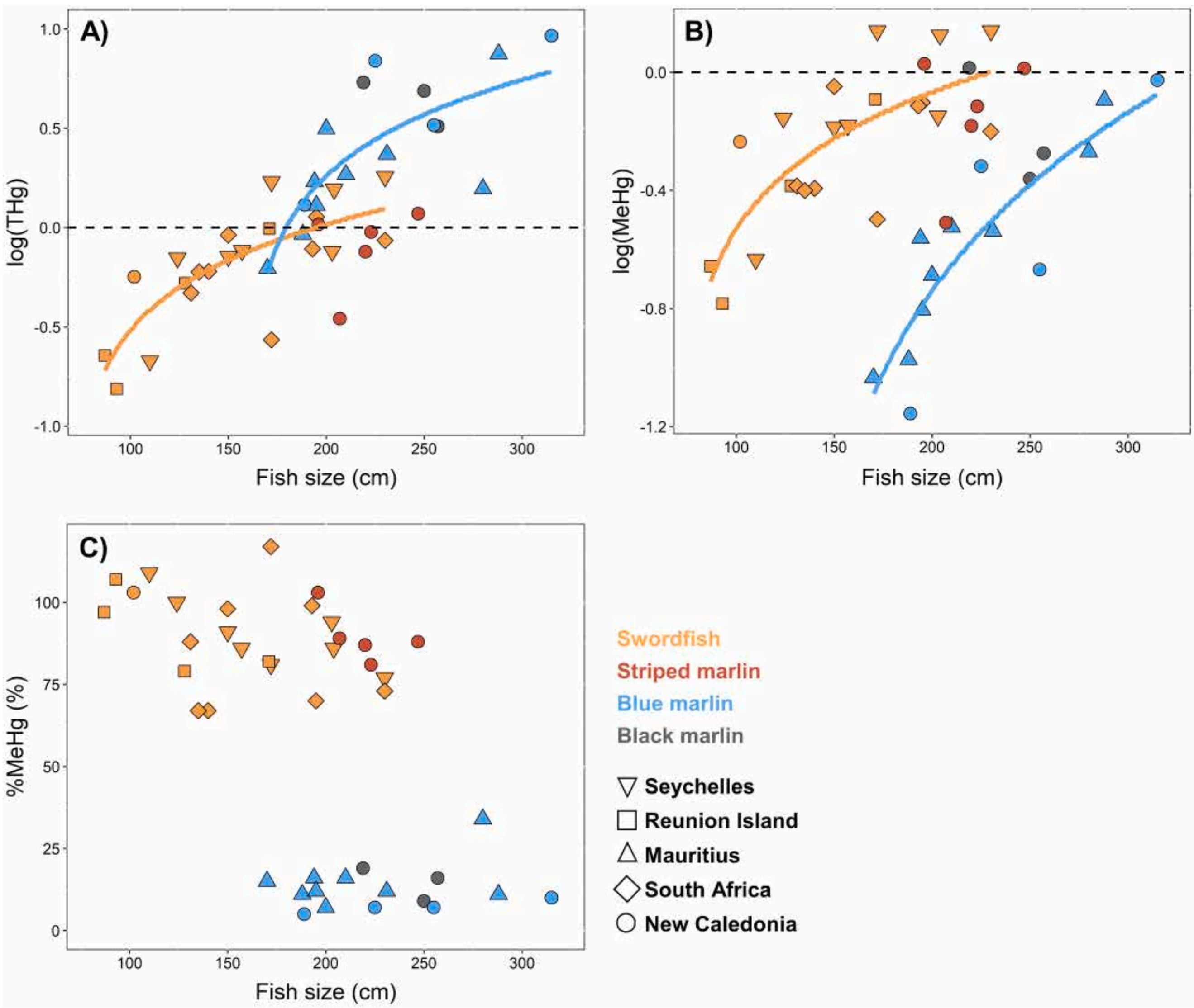

Fig. 3. Investigating the potential effects of species, ocean region, and fish size, on mercury and methylmercury content in billfish muscles. A), B), and **C)** show the relationship between log-transformed total mercury (THg) concentrations, log-transformed methylmercury (MeHg) concentrations, and percentage of MeHg (%MeHg) in total Hg, respectively, according to fish size (lower jaw fork length LJFL, i.e., from the tip of the lower jaw to the fork of the tail). Colours and shapes of the symbols refer to species and samples provenance, respectively. The dashed lines in panels A) and B) correspond to the MeHg guideline value of 1 μg.g^{-1} ww (i.e. 0 after log-transformation) (FAO and WHO, 2005). The curves in panels A) and B) illustrate the bioaccumulation curves fitted between log-transformed Hg concentrations and fish size for swordfish (THg: $R^2 = 0.60$, $log(THg) = 4.47 \times (LJFL - 71.80^{0.06}) - 6.00$; MeHg: $R^2 = 0.58$, $log(MeHg) = 4.54 \times (LJFL - 70.77^{0.05}) - 6.00$) and blue marlin (THg: $R^2 = 0.68$, $log(THg) = 5.02 \times (LJFL - 159.54^{0.06}) - 6.00$; MeHg: $R^2 = 0.78$, $log(MeHg) = 3.14 \times (LJFL - 110.58^{0.12}) - 6.00$), combining Hg data from all ocean regions as no regional differences were found in our data (Kruskal-Wallis, $p > 0.05$ for the two species). (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

documented for MeHg in marine organisms (e.g., Branco et al., 2007; Kojadinovic et al., 2006; Shultz and Crear, 1976; Vega-Sánchez et al., 2017), mostly by forming complexes with cysteine (MeHgCys) (Amlund et al., 2007; Leaner and Mason, 2004). It is also observed for inorganic Hg in fish species through the probable binding to proteins in muscle tissues as well (Manceau et al., 2021a). Conversely, species and ocean regions were not significant parameters explaining observed differences of total Hg concentrations in our dataset, suggesting no inter-species and regional differences of total Hg concentrations among the four billfish species in the western Indian and the southwestern Pacific, once accounting for differences in fish sizes (Fig. 3A). Yet, given our small sample size, a larger billfish Hg dataset covering a wide range of billfish sizes would be needed to fully investigate these aspects. Individual total lipid content was not a significant parameter to explain total Hg variability, which does not support the general hypothesis that the fatter the fish, the lower its total Hg concentration (Balshaw et al., 2008).

Methylmercury concentrations in billfishes displayed lower variability than total Hg concentrations, with the lowest value (0.07 μg.g^{-1} ww) measured in blue marlin from New Caledonia, and the highest one (1.38 μg.g^{-1} ww) in a swordfish from Seychelles (Table 1). Methylmercury concentrations in the present study are similar to the scarce MeHg levels reported in the four species from different regions of the global ocean (SI Appendix Table S1). Variability of MeHg concentrations was mainly explained by fish size and species, but neither by ocean region nor individual lipid content (Fig. 3B; ANCOVA, $p < 0.05$ for both fish size and species, p-value > 0.05 for other explanatory variables). These results once again highlight the natural process of MeHg bioaccumulation in individuals through time, but also suggest inter-species differences in MeHg biomagnification and toxicokinetics.

3.2. Mercury speciation patterns across billfish species

To investigate differences of MeHg biomagnification processes, we compared %MeHg measured in muscle tissues of billfishes. Variability of %MeHg was explained by species, but not by fish size, ocean region or lipid content (ANCOVA, $p < 0.05$ for species, $p > 0.05$ for other explanatory variables), with swordfish (89 ± 14 %, all regions combined) and striped marlin (90 ± 8 %) exhibiting significantly higher %MeHg than blue (12 ± 7 %) and black (15 ± 5 %) marlins (Table 1; Fig. 3C). Low %MeHg in blue marlin is similar to values previously reported in this species from French Polynesia (19 ± 22 %; Manceau et al., 2021) and Hawaii (mean %MeHg values between 13 and 29 %; Rivers et al., 1972; Shultz et al., 1976; Shultz and Crear, 1976; Shultz and Ito, 1979), but is lower than observations from the Atlantic Ocean (43 ± 3 %; Yamashita et al., 2005). Percentage of MeHg values measured in our black marlin samples are much lower than those, albeit limited, reported in the literature (68 ± 13 %; Kacprzak and Chvojka, 1976). High %MeHg values in swordfish and striped marlin measured in the present study are similar (Branco et al., 2007; Kamps et al., 1972) or slightly higher (Brambilla et al., 2013; Chen et al., 2007; Forsyth et al., 2004; Yamashita et al., 2005) than the ones available in the literature from different ocean regions (SI Appendix Table S1). Low %MeHg in blue and black marlins suggest these two species are capable of *in vivo* demethylation, as already evidenced in blue marlin from French Polynesia and Hawaii in which %MeHg were found to decrease with increasing total Hg concentrations (SI Appendix Fig. S1) (Manceau et al., 2021a; Shultz et al., 1976; Shultz and Crear, 1976). Here, we found no significant negative power-law relationship between %MeHg and total Hg concentrations in blue and black marlins samples, which might be explained by the lack of small individuals in our dataset (all our Hg data were obtained in individuals ≥ 170 cm; SI Appendix Fig. S1), which decreases our ability to detect the trend.

In blue marlin muscle tissue, MeHg demethylation is supposed to rely on the strong affinity of inorganic Hg for Se, with HgSe nanoparticles likely being the final detoxification product (Manceau et al., 2021a). To further investigate the possible role of Se in MeHg demethylation, we explored the relationship between inorganic Hg and total Se molar concentrations in billfish muscle tissues (Fig. 4). There was no significant linear relationship between iHg and Se in striped marlin and swordfish (p-value > 0.05). Conversely, in blue marlin, inorganic Hg molar concentrations increased significantly and positively with Se molar concentrations (p-value < 0.05, $R^2 = 0.98$) with a ~ 1:1 molar ratio. This suggests that almost all inorganic Hg is associated to Se in blue marlin muscle, which agrees with a Se-mediated MeHg demethylation pathway in the muscle tissue itself for this species. Overall, further characterization of Hg and Se speciation in different tissues of billfish species that do (e.g., blue and black marlin) and do not (e.g., swordfish and striped marlin) demethylate MeHg would be needed to better understand possible *in vivo* MeHg detoxification pathways in these top marine predators. Among Se forms, selenoneine in particular would deserve being quantified in billfish muscle as it is suspected to be responsible for MeHg demethylation and excretion in marine organisms (El Hanafi et al., 2022; Yamashita et al., 2013).

4. Toxicological assessment of billfish consumption

About 13 % of our billfish samples (6 over 45) displayed MeHg concentrations above the fish consumption guideline value of 1 μg.g^{-1} ww, corresponding to three swordfish individuals from Seychelles, and two striped marlin and one black marlin from New Caledonia (Fig. 3B). This number rises up to 50 % (23 over 45) if we compare total Hg concentrations to the MeHg guideline value (Fig. 3A), as commonly done when evaluating the potential risks to human health of billfish consumption (e.g., Bille et al., 2020; Kojadinovic et al., 2006; Soto-Jiménez et al., 2010). This relies on different species-specific MeHg biomagnification patterns, especially the likely *in vivo* demethylation capability of blue and black marlins in muscle tissue, and highlights the importance of considering MeHg concentrations, rather than total Hg concentrations, when assessing the risks associated to billfish consumption. Alternatively, species-specific conversion factors between total and MeHg concentrations, defined on a sufficient number of samples, could be used to evaluate the potential risks of billfish consumption in regions with limited or no access to advanced and costly laboratory facilities needed to measure MeHg concentrations.

To further explore the potential risks to human health of billfish consumption, we estimated the maximum recommended number of fish meals per week, using a PTWI value of 1.3 μg MeHg/kg bw (EFSA, 2012) and considering mean fish portions of 150 g and 75 g for adults and children, respectively. In our specific dataset, MeHg concentrations in flesh indicate that about half of the fish samples should only be

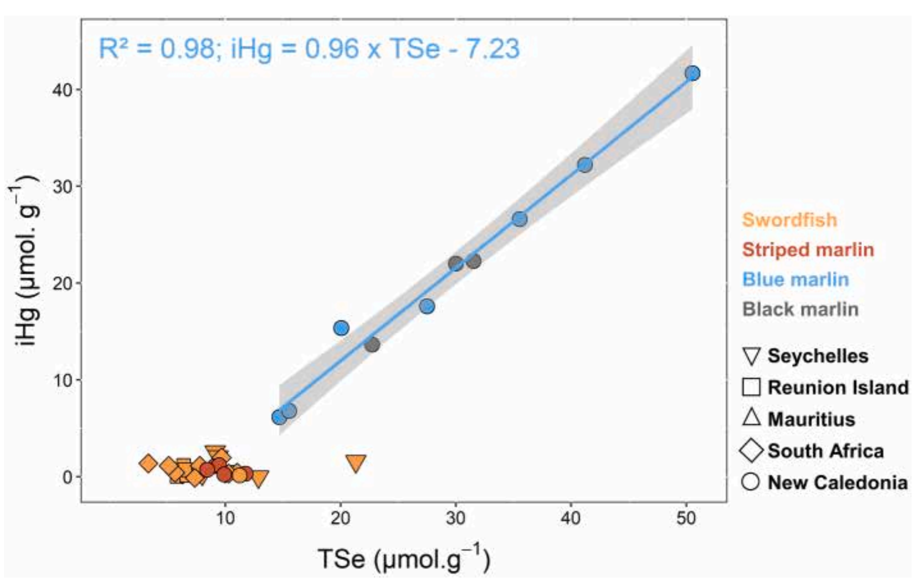

Fig. 4. Relationship between inorganic mercury (iHg) and total selenium (TSe) molar concentrations (μmol.g^{-1}) in billfish muscle samples. Colours and shapes of the symbols refer to species and samples provenance, respectively. The blue line and the grey shadow represent the significant linear regression between the two variables found for blue marlin only ($p < 0.05$; $p > 0.05$ for striped marlin and swordfish; not tested for black marlin because of insufficient sample availability), and the standard deviation, respectively. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

consumed occasionally (i.e., less than once a week) by children (52 % of the fish samples) and adults (45 %) (Fig. 5A & B). This concerns mostly swordfish (n = 13 and 12 over the 21 fish, for children and adults, respectively) and striped marlin (n = 4 over the 5 fish, for both people categories) because of their high %MeHg. Conversely, most of the samples of blue and black marlins (> 80 %, both species combined) could be consumed once a week, or even more frequently, by both children and adults because of their low %MeHg. These results highlight once again the need to adapt dietary advice to each billfish species and nuances the general idea that billfish should be avoided for consumption because of their high MeHg levels. Here, the toxicological assessment of billfish consumption relied on the assumption that inorganic Hg in blue and black marlins muscle is mostly complexed as inert and poorly bioavailable HgSe particles, as found by Manceau et al., (2021a), and suggested by the strong correlation between inorganic Hg and total Se in our dataset. Yet, this should be verified with additional characterization of Hg complexation, bioavailability, gastro-intestinal absorption and toxicity in the different billfish species. Furthermore, this toxicological assessment assumes that weekly MeHg exposure occurs solely through the consumption of billfishes, whereas other dietary sources of MeHg might also play a significant role. In coastal regions in particular, where fishes are central for local subsistence, the consumption of other oceanic species with quite high MeHg concentrations relative to the rest of the food web (e.g., tropical tunas and wahoo), and of coastal species with lower MeHg concentrations (generally below 0.3 $\mu g.g^{-1}$ ww; e.g., parrotfish, bream) (ANSES, 2019), represents an additional source of MeHg that should be considered when developing dietary recommendations.

In addition to species and MeHg metabolism, individual fish body size itself influenced significantly weekly recommendations for billfish consumption. For all species, regardless of their %MeHg, the maximum recommended number of fish meals per week decreased with increasing fish size (Fig. 5), as MeHg concentrations tend to increase with fish body size. This illustrates the general principle that the bigger the fish, the lower the number of meals recommended, and complicates the definition of weekly consumption guidelines per species currently used by food agencies. Fish body size mostly depends on the fishing gears that select preferentially certain billfish sizes, and on ocean regions (IOTC Secretariat, 2022). To date, fish size or weight ranges and fishing area are rarely reported in Hg exposure assessments (e.g., ANSES, 2019; Bille et al., 2020). Providing such information for a globalized fishing industry (i.e., fish consumed in a country can originate from different regions of the global ocean) is challenging, but can be assessed more easily in tropical coastal communities where billfish consumption mostly relies on semi-industrial and recreational coastal catches. In these areas, Hg risk assessments associated with billfish consumption should rely on MeHg concentrations measured in a large number of samples representing the diversity of fishing gears and catch regions, and covering a large range of fish size. In addition, a general recommendation could be to avoid regularly consuming the biggest specimens. Based on our specific dataset and species-specific MeHg concentrations, consumers should for example avoid frequently eating blue and black marlin specimens bigger than 250 cm, and swordfish and striped marlins bigger than 150 cm (Fig. 5). These fish size thresholds are only preliminary, and a more extensive Hg dataset covering a broader range of billfish sizes is needed to reliably determine the fish sizes warranting avoidance.

Dietary advices of billfish consumption would also benefit from the complementary characterization of essential fatty acids, proteins, micronutrients, and minerals in billfish muscle, given their role in sustaining food security in coastal regions and for human nutrition benefits (FAO/WHO, 2011; Swanson et al., 2012). On the other hand, evaluation of legacy and emerging persistent organic pollutants (POPs) would be useful to strengthen the consumption recommendations, given their toxicity and accumulation in marine food webs. A wide range of POPs were measured in swordfish in the Seychelles archipelago and along the Brazilian coasts, revealing the widespread presence of anthropogenic pollutants in the southern oceans (de Azevedo e Silva et al., 2007; Munschy et al., 2023, 2020a, Munschy et al., 2020b), and calling for more systematic POPs monitoring in highly consumed tropical top predators. Combining all these compounds in benefit-risk assessments would be valuable to refine which billfish species could help reducing food insecurity in tropical coastal communities (Sabino et al., 2022; Sardenne et al., 2020; Wang et al., 2019). Finally, as cooking temperature is suspected to impact Hg concentrations, complexation and bioavailability in fish muscle (Afonso et al., 2015; Amyot et al., 2023), further characterisation of Hg concentrations and complexation in

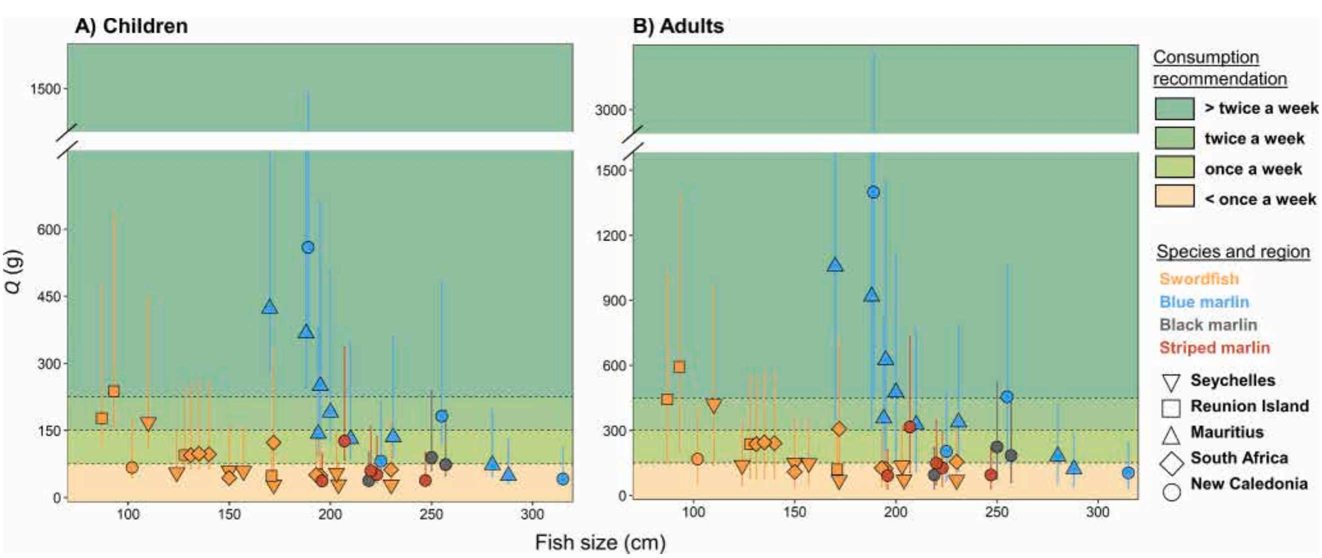

Fig. 5. Influence of fish species, fish body size, consumer body weight, and mercury speciation on billfish consumption recommendations. The quantity of fish (Q, g) at which Provisional Tolerable Weekly Intake (PTWI) of methylmercury (MeHg) is reached (i.e., above which consumption may pose health problems regarding MeHg intake) was compared to mean fish portions (75 g and 150 g for children and adults, respectively) to estimate the maximum recommended number of fish meals per week for **A)** children and **B)** adults. Colored horizontal bands symbolised the weekly dietary advices: more than twice a week, twice a week, once a week, and less than once a week (i.e. occasionally), from darker green to yellow, respectively. Colours of the symbols refer to species while shapes indicate sample provenance. Symbols show the mean values of Q and lines their standard deviations, considering 30 ± 10 kg and 75 ± 25 kg body weights, for children and adults, respectively. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

different frequently consumed billfish products (e.g., raw, boiled, grilled, smoked and fried) would be needed to refine risk assessment (which considers that fish is eaten raw) associated with actual billfish consumption habits.

5. Conclusion

With the adoption of the Minamata Convention, it has become a priority to document Hg content in marine commercial species, and to assess MeHg exposure to humans through seafood consumption. This is of particular importance in coastal regions and/or small islands where fishes are central for local subsistence, nutrition, culture, and economics, and where populations are thus exposed to potentially high MeHg doses (Basu et al., 2018). Here, the investigation of total Hg and MeHg content in swordfish and three marlin species sheds light on different species-specific MeHg biomagnification patterns, likely related to different capabilities of *in vivo* demethylation in muscle tissue involving Se. These results call for further characterization of Hg and Se complexation in the four studied species to better understand the mechanisms responsible for MeHg detoxification. In relation to these different species-specific MeHg biomagnification patterns, we found that children and adults can eat more frequently blue and black marlins than swordfish and striped marlin, which highlights the need to adapt dietary advices to each billfish species, instead of considering them as a single fish category, as commonly done.

CRediT authorship contribution statement

Anaïs Médieu: Writing – original draft, Methodology, Investigation, Formal analysis, Conceptualization. **David Point:** Writing – review & editing, Supervision, Investigation, Formal analysis. **Valérie Allain:** Writing – review & editing, Resources, Investigation, Funding acquisition. **Nathalie Bodin:** Writing – review & editing, Resources. **Mélanie Lemire:** Writing – review & editing, Investigation. **Pierre Ayotte:** Writing – review & editing, Investigation. **Zahirah Dhurmeea:** Writing – review & editing, Resources. **Matthieu Waeles:** Writing – review & editing, Formal analysis. **Laure Laffont:** Writing – review & editing, Formal analysis. **Antoine Le Gohalen:** Writing – review & editing, Formal analysis. **François Roupsard:** Writing – review & editing, Resources. **Anne Lorrain:** Writing – review & editing, Supervision, Investigation, Funding acquisition.

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.

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Appendix A. Supplementary material

Supplementary data to this article can be found online at https://doi.org/10.1016/j.envint.2025.109252.

Data availability

Data will be made available on request.

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