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# Concentrations and stable isotopes of mercury in sharks of the Galapagos Marine Reserve: Human health concerns and feeding patterns

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## ABSTRACT

The human ingestion of mercury (Hg) from sea food is of big concern worldwide due to adverse health effects, and more specifically if shark consumption constitutes a regular part of the human diet. In this study, the total mercury (THg) concentration in muscle tissue were determined in six sympatric shark species found in a fishing vessel seized in the Galapagos Marine Reserve in 2017. The THg concentrations in shark muscle samples ($n = 73$) varied from 0.73 mg kg$^{-1}$ in bigeye thresher sharks (*Alopias superciliosus*) to 8.29 mg kg$^{-1}$ in silky sharks (*Carcharhinus falciformis*). A typical pattern of Hg bioaccumulation was observed for all shark species, with significant correlation between THg concentration and shark size for bigeye thresher sharks, pelagic thresher sharks (*Alopias pelagicus*) and silky sharks. Regarding human health concerns, the THg mean concentration exceeded the maximum weekly intake fish serving in all the studied species. Mass-Dependent Fractionation (MDF, $\delta^{202}$Hg values) and Mass-Independent Fractionation (MIF, $\Delta^{199}$Hg values) of Hg in whitetip sharks (*Carcharhinus longimanus*) and silky sharks, ranged from 0.70‰ to 1.08‰, and from 1.97‰ to 2.89‰, respectively. These high values suggest that both species are feeding in the epipelagic zone (i.e. upper 200 m of the water column). While, blue sharks (*Prionace glauca*), scalloped hammerhead sharks (*Shyrna lewini*) and thresher sharks were characterized by lower $\Delta^{199}$Hg and $\delta^{202}$Hg values, indicating that these species may focus their foraging behavior on prey of mesopelagic zone (i.e. between 200 and 1000 m depth). In conclusion, the determination of THg concentration provides straight-forward evidence of the human health risks associated with shark consumption, while mercury isotopic compositions constitute a powerful tool to trace the foraging strategies of these marine predators.

*Capsule:* A double approach combining Hg concentrations with stable isotopes ratios allowed to assess ontogeny in common shark species in the area of the Galapagos Marine Reserve and the human health risks concern associated to their consumption.

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## 1. Introduction

Mercury (Hg) contamination in the environment produces considerable adverse effects on marine ecosystems, as well as on human health (Zheng et al., 2019; US EPA, 2018). Mercury is known for being a highly toxic pollutant once it is methylated to monomethyl-Hg (MMHg) and dimethyl-Hg by abiotic and biotic processes (Sunderland et al., 2009). The main sources of Hg in the Ocean are the rivers inputs through estuaries, submarine volcanic activity, and atmospheric depositions (Obrist et al., 2018). Most of the Hg in the atmosphere and surface ocean results from human activities, particularly from coal-fired power stations, metallurgy, and artisanal gold mining, Hg production and use, and industrial metals extraction (Obrist et al., 2018; Streets et al., 2019). These processes release important amounts of inorganic Hg (iHg) in gaseous ($Hg^0$) or dissolved forms, as $Hg^{+2}$ (Bergquist and Blum, 2007). The oxidation and reduction of these chemical Hg species occur simultaneously by photochemical activation. In aquatic systems, the presence of organic matter also influences Hg photoreduction (O'Driscoll et al., 2008). The main oxidation and reduction mechanisms in the aquatic environment are, therefore, induced by photochemical and dark biotic processes (Fitzgerald et al., 2007; Sunderland et al., 2009).

Methyl-Hg biomagnification along the trophic web results in elevated Hg concentrations in top predators (Mason et al., 1995; Chen et al., 2014). Moreover, as long-lived species, sharks usually display significant MMHg concentrations due to bioaccumulation and bioamplification processes (McKinney et al., 2016). Concentration of MMHg in fish depends on their trophic level, size (age), and environment; therefore, the highest MMHg concentrations are generally found in large carnivorous fish (Maurice-Bourgoin et al., 2000; Fitzgerald et al., 2007). Large pelagic fish (including sharks) are top predators, and their diet is generally composed of high trophic level fish or squids.

Monomethyl-Hg is a neurotoxic molecule; it can affect the development of young children exposed *in utero* or during the early childhood (Grandjean et al., 1997; Mergler et al., 2007). The ingestion of Hg and, more specifically MMHg can cause neurological problems, deficits in motor, psychomotor, visual and /or cognitive functions, immune deficiencies and toxicity to the central nervous system (You et al., 2012; Kim et al., 2016). Consumption of large pelagic fish appears to be the major pathway of human exposure to MMHg (Morel et al., 1998; Dorea and Barbosa, 2003). Therefore, from the human health perspective, it is crucial to document Hg levels in predator fish of the Pacific region and to understand which food items tend to increase the risk of Hg exposure.

From 1990–2018, according to the FAO (2020), the global capture fisheries increased by 14% and reached 96.4 million tons in 2018, which 90% was caught in the oceans. Asian countries represent almost half of the global marine capture fisheries, averaging 45 million tons per year (FAO, 2020). Global shark catches reported to FAO (2019) have tripled since 1950, reaching an all-time high in 2000 with 868,000 tons. Since then, a downward trend can be observed with about 22% lower catches (680,000 tons) in 2018. Asian fishing countries only report non-identified catches of sharks and rays or do not report any statistics at all for this group (FAO, 2020). In China, experts estimate that annual shark fishing ranges between 10 and 15,000 tons and almost all is domestically consumed (FAO, 2020). Nonetheless, one of the biggest gaps in the discussion of shark fin and shark meat markets is the lack on data to estimate the annual consumption in Asia (Dent and Clarke, 2015). Among species that are usually targeted, are the silky shark (*Carcharhinus falciformis*) and the oceanic white tip (*Carcharhinus longimanus*), while blue shark (*Prionace glauca*) is mainly caught as bycatch (Vannuccini, 1999).

Shark fin is one of Asia's most valued and luxury seafood products (Garcia Barcia et al., 2020). These species are consumed primarily for their meat, skin, cartilage liver, and fins, which are highly appreciated in Asiatic culture for the traditional shark fin soup. The meat of some shark species (i.e., thresher sharks) is considered of higher quality than others. Nevertheless, pelagic thresher shark (*Alopias pelagicus*) and bigeye thresher shark (*Alopias superciliosus*) meat is judged of lower quality compared to that of common thresher shark (*Alopias vulpinus*), but it is also widely commercialized (Vannuccini, 1999). Blue shark is considered as one of the least preferred species for human consumption due to its soft and strong flavored meat; this species is frequently caught as bycatch, but is usually discarded, often after finning (FAO, 2020). Blue shark has a limited market in France, Germany, Spain, and Italy. In Japan, blue sharks are used for the preparation of *hanpen* (shark paste), but only if they have been promptly processed after capture. Large shark species are often avoided for human consumption as they can accumulate high levels of mercury and other toxic metals (Vannuccini, 1999). Concentrations of about 0.5–1 μg Hg $g^{-1}$ wet weight (w.w.) in axial muscle (>90% of Hg in muscle is MMHg) produce several changes in fish, including alteration in biochemical processes, damage to cells and tissues, and reduced reproduction (Scheuhammer et al., 2014).

In addition to carbon and nitrogen stable isotopes analysis ($\delta^{13}C$ and $\delta\ ^{15}N$) to characterize dietary resources and the trophic level, Hg stable isotopes are increasingly used in environmental studies to connect geochemistry, biology, ecology (Le Croizier et al., 2020, Tsui et al., 2020) and health sciences. Mercury stable isotopes present multiple useful signatures due to classical Mass-Dependent Fractionation (MDF, reported as $\delta^{202}Hg$) and unique photochemical Mass-Independent Fractionation (MIF, reported as $\Delta^{199}Hg$). These properties enable tracing MMHg sources in marine and freshwaters environments (Cransveld et al., 2017; Masbou et al., 2018; Sackett et al., 2015; Laffont et al., 2009). Hg MDF is the result of various abiotic, such as photoreduction and volatilization (Bergquist and Blum, 2007; Zheng et al., 2007) and biotic processes, the most important being methylation and demethylation (Janssen et al., 2016; Perrot et al., 2016). Hg MIF occurs predominantly during photochemical reactions (Bergquist and Blum, 2007). In seawater, solar radiations induce a Hg isotopic gradient from the surface to depth, with higher $\delta^{202}Hg$ and $\Delta^{199}Hg$ values in the epipelagic zone (between 0 and 200 m deep) than in the mesopelagic zone (between 200 and 1000 m deep) (Blum et al., 2013; Sackett et al., 2017). Therefore, Hg isotopic signatures constitute a powerful tool to trace the feeding patterns of marine predators, for instance discriminating epipelagic from mesopelagic foraging habitats (Madigan et al., 2018; Le Croizier et al., 2020) as both $\delta^{202}Hg$ and $\Delta^{199}Hg$ values decrease with depth in pelagic consumers (Blum et al., 2013; Motta et al., 2019).

In August 2017, a fishing vessel (Fu Yuan Yu Leng 999) was seized in the Galapagos Marine Reserve, and hundreds of sharks have been sampled for scientific purpose. In order to document the Hg levels of the most common shark species living in the Eastern Equatorial Pacific, we present total mercury concentrations (THg) in muscle tissue of six sympatric shark species and combine these results with the Hg isotope signatures to provide information on the biology and feeding patterns of these shark species. To assess the health risk associated with a regular shark food consumption, we also calculated the maximum weekly tolerable amount of shark meat by fish species, for adults and children.

## 2. Materials and methods

### 2.1. Sampling

This research was performed under Galapagos National Park Directorate (GNPD) research permit PC-24-17 and was carried out following the protocols of ethics approved by Ecuadorian laws. Sampling was done on board a Chinese refrigerated cargo vessel "Fu Yuan Yu Leng 999" boarded in August 2017 by the GNPD and the Ecuadorian Navy while illegally navigating within the Galapagos Marine Reserve, in the Eastern Equatorial Pacific. Within the ship, 6 623 shark carcasses from several protected species were found (Alava et al., 2017). From the six captured shark species, one, the hammerhead shark (*Sphyrna lewini)*, is classified as endangered by the IUCN, one near threatened (blue shark *Prionace glauca*), and four vulnerable (Oceanic whitetip shark *Carcharhinus* *longimanus,* silky shark *Carcharhinus falciformis,* bigeye thresher *Alopias superciliosus,* and pelagic thresher *Alopias pelagicus*).

A total of 73 sharks (14 scalloped hammerhead, 12 blue shark, 14 oceanic whitetip, 13 silky shark, 6 big eye thresher shark and 14 pelagic thresher shark) were randomly sampled and preserved frozen (−18 °C). Weight (kg) and size (cm), defined as the pre-caudal length (PCL) thereafter in the text, were recorded for all animals (Table 1). All samples for mercury analysis were homogenously selected within the whole ranges of PCL and weight (Fig. SI_1).

### 2.2. Sample preparation

Original samples were composed of muscle tissue and skin and were cut directly from the shark body, placed in separate zip-lock bags and frozen for preservation before their transport to the laboratory. Once in the Environmental Engineering Laboratory at the Universidad San Francisco de Quito (LIA-USFQ, Ecuador), samples were thawed, skin was removed from the muscle tissue with a ceramic knife to reduce the risk of metal contamination, and almost one hundred grams of muscle were kept in zip-lock bags at 4 °C. Samples had no contact with human dermal layer nor metal surfaces during their manipulation.

Approximately 20 g from the samples were freeze-dried at −50 °C, at the Laboratory of Agricultural and Food Biotechnology (USFQ) and the moisture content was 75%. Dried samples were grounded and homogenized before their storage in new zip-lock bags.

### 2.3. Total mercury (THg) analysis

As total mercury (THg) concentration is known to be almost exclusively in the MeHg form in shark muscle, including for the species analyzed here, e.g. 95–98% in blue sharks (Carvalho et al., 2014; Kim et al., 2016; Storelli et al., 2003), THg was used as a proxy for MeHg. All samples were processed within one month using a DUAL-cell DMA-80 Direct Mercury Analyzer (Milestone Inc.), based on the EPA 7374 analytical method by thermal desorption at the Universidad San Francisco de Quito (LIA-USFQ, Ecuador). Samples did not require pretreatment or acid-digestion; approximately 15 mg of sample was weighed and placed into the equipment to be pyrolyzed at 800 °C and analyzed in duplicate or triplicate to ensure precise results. Reference material CRM DORM-4 was used to control the accuracy of the procedure. Average relative error reached 10% and CRM recovery 98%. The values obtained were expressed in milligrams per kilogram on a dry weight basis (d.w.) and were converted to wet weight, considering the loss of water of muscle tissue during the freeze-drying process. The same conversion factor of 4 (from wet to dry weight) in shark muscles has been used by several authors (Pethybridge et al., 2010; Bosch et al., 2013).

The THg body mass index (BMI) was calculated for each shark sample as THg ($mg.kg^{-1}$) per kg of body mass (w.w.) according to Pethybridge and collaborators (2010).

### 2.4. Hg stable isotopes analysis

A subset of 31 shark tissues (i.e. 5 or 6 individuals covering the available size range for each species) was selected for Hg isotope analysis. Aliquots of approximately 20 mg of dry shark muscle were left overnight at room temperature in 3 mL of concentrated bi-distilled nitric acid ($HNO_3$). Samples were then digested on a hotplate for 6 h at 85 °C in pyrolyzed glass vessels closed by Teflon caps. One mL of hydrogen peroxide ($H_2O_2$) was added and digestion was continued for another 6 h at 85 °C. One hundred μL of BrCl was then added to ensure a full conversion of methylmercury to inorganic Hg. The digest mixtures were finally diluted in an inverse aqua regia solution (3 $HNO_3$: 1 HCl, 20 vol% MilliQ water) to reach a nominal Hg concentration of 1 ng $g^{-1}$. Aliquots of certified reference materials (ERM-BCR-464, tuna flesh) and blanks were prepared in the same way as the tissue samples.

Mercury isotope compositions were measured at the Observatoire Midi-Pyrenées (OMP, France) using multi-collector inductively coupled plasma mass spectrometry (MC−ICP−MS, Thermo Finnigan Neptune Plus) with continuous-flow cold vapor (CV) generation using Sn(II) reduction (CETAC HGX-200), according to a previously published method (Enrico et al., 2016; Goix et al., 2019). Hg isotope composition is expressed in δ notation and reported in parts per thousand (‰) deviation from the NIST SRM 3133 standard, which was determined by sample-standard bracketing according to the following equation:

$$\delta^{202}Hg\ (‰) = [\ (^{202}Hg/^{198}Hg)_{sample}\ /\ (^{202}Hg/^{198}Hg)_{NIST\ 3133})\ -1]\ *\ 1000$$

Where, $\delta^{202}$ represents the mass of $^{202}Hg$ isotope. $\delta^{202}Hg$ is used as a measure of MDF. Measures of MIF are calculated as the difference between a measured δ-value, and the predicted δ-value that is calculated by multiplying the measured $\delta^{202}Hg$ value by the kinetic MDF fractionation factor for each isotope (Bergquist and Blum, 2007). Δ notation is used to express Hg MIF by the following equation:

$$\Delta^{199}Hg\ (‰) = \delta^{202}Hg - (\delta^{202}Hg\ Xa)$$

Total Hg in the diluted digest mixtures was monitored by MC-ICP-MS using $^{202}Hg$ signals: mean recoveries of 103 ± 5% (1σ, n = 31) for samples and 100 ± 2% (1σ, n = 5) for certified reference materials were found, ensuring efficient digestion of samples. Hg levels in blanks were below the detection limit. Reproducibility of Hg isotope measurements was assessed by analyzing UM-Almadén (n = 5), ETH-Fluka (n = 5) and the biological tissue procedural standard ERM-BCR-464 (n = 5). Only one analysis was performed per sample, but measured isotope signatures

**Table 1**
Common and scientific names, number of samples (n), IUCN status, measured pre-caudal length range (average), measured body mass range (average), total mercury concentrations (THg, mg $kg^{-1}$ d.w. and w.w.), THg body mass index (BMI) calculated as THg per kg of body mass (Pethybridge et al., 2010), MDF ($\delta^{202}Hg$) and MIF ($\Delta^{199}Hg$) range (and average) for the six shark study species samples from the Eastern Equatorial Pacific.

| Common name | Scientific name | n | IUCN status | Pre-caudal length range (cm) (Average) | Body mass range (kg) (Average) | THg (mg $kg^{-1}$) d.w. | THg (mg $kg^{-1}$) w.w. | THg BMI | $\delta^{202}Hg$ (‰) | $\Delta^{199}Hg$ (‰) |
|---|---|---|---|---|---|---|---|---|---|---|
| Scalloped hammerhead | *Sphyrna lewini* | 14 | Endangered | 109–213 (147) | 11–94 (38) | 1,17–19,01 (7,11) | 0,29–4,75 (1,78) | 0,01–0,10 (0,05) | 0,46–0,67 (0,56) | 1,54–2,19 (1,87) |
| Blue shark | *Prionace glauca* | 12 | Near threatened | 100–149 (120) | 5–29 (12) | 3,69–13,41 (7,50) | 0,92–3,35 (1,88) | 0,07–0,36 (0,18) | 0,45–0,73 (0,57) | 1,70–2,24 (1,88) |
| Oceanic whitetip | *Carcharhinus longimanus* | 14 | Vulnerable | 68–120 (101) | 4–18 (12) | 1,99–15,58 (5,21) | 0,50–3,90 (1,30) | 0,04–0,33 (0,12) | 0,84–1,08 (0,94) | 2,43–2,86 (2,69) |
| Silky shark | *Carcharhinus falciformis* | 13 | Vulnerable | 56–166 (107) | 1–45 (15) | 1,01–33,58 (7,05) | 0,25–8,39 (1,76) | 0,02–0,29 (0,11) | 0,70–0,96 (0,81) | 1,97–2,89 (2,49) |
| Bigeye thresher shark | *Alopias superciliosus* | 6 | Vulnerable | 114–182 (151) | 21–69 (49) | 2,98–10,23 (6,04) | 0,75–2,56 (1,51) | 0,02–0,04 (0,03) | 0,10–0,78 (0,48) | 1,49–2,45 (1,80) |
| Pelagic thresher | *Alopias pelagicus* | 14 | vulnerable | 63–174 (126) | 5–69 (28) | 0,74–7,24 (3,15) | 0,18–1,81 (0,79) | 0,02–0,05 (0,03) | 0,10–0,83 (0,45) | 1,43–2,91 (1,94) |

as well as analytical reproducibility of standards agreed with previously published values (Table SI_1; Blum et al., 2013; Jiskra et al., 2017; Li et al., 2016; Masbou et al., 2013).

### 2.5. Human health risk assessment

Considering a Provisional Tolerable Weekly Intake (PTWI) of 1.6 µg MMHg kg$^{-1}$ Human Body Weight (HBW) proposed by the World Health Organization (WHO, 2018), we calculated the maximum weekly intake of shark (g shark weekly) according to the following equation:

$$Intake = \frac{PTWI * HBM}{THg}$$

Where: THg represents the average THg concentration (mg kg$^{-1}$ w.w.) in each shark species measured in this study (Table 1). Human body mass (HBM) was equal to 70 kg, 60 kg and 15 kg for adult men, women, and children, respectively.

### 2.6. Data analyses

For comparison of Hg concentration and Hg isotope ratios between species, data were first assessed for normality (Shapiro–Wilk tests) and homogeneity of variances (Bartlett tests). As these conditions were not met, Kruskal–Wallis (KW) tests, followed by Conover–Iman multiple comparison tests with Bonferroni's adjustment were used. Linear regressions were conducted to assess the relationship between Hg concentration and shark length. Analysis of covariance, ANCOVA, was used to compare the Hg accumulation rates between species.

## 3. Results and discussion

### 3.1. Levels of THg: influence of shark size and species

Total Hg concentrations in the muscles of the 6 studied shark species ranged between 0.18 mg kg$^{-1}$ w.w. in *Alopias pelagicus* and 8.29 mg kg$^{-1}$ w.w. in *Carcharhinus longimanus*, corresponding to 0.72–33.16 mg kg$^{-1}$ d.w, respectively (Fig. 1A; Table 1). *Prionace glauca* showed the highest mean THg concentration with a value of $1.88 \pm 0.63$ mg kg$^{-1}$ w.w. ($n = 12$), while the lowest values were measured in *Alopias pelagicus* with $0.79 \pm 0.47$ mg kg$^{-1}$ w.w. ($n = 14$) (Fig. 1A; Table 1). THg concentrations were significantly higher in *A. superciliosus* and *P. glauca* compared to *A. pelagicus* and *C. falciformis* (KW, $p < 0.05$). The diet biomass of *P. glauca* is largely dominated by cephalopods (98%), while this proportion is lower in the other species (Galván-Magaña et al., 2013). Moreover, the two species of thresher sharks analyzed here target different fish prey, with *A. pelagicus* focusing primarily on lanternfish and *A. superciliosus* on hakes (Galván-Magaña et al., 2013). Although trophic competition has been previously suggested for these predator species (Páez-Rosas et al., 2018), the differences in prey consumed may partly explain the differences in THg concentrations between shark species.

Pre-caudal length of the sharks ranged from 56 to 203 cm and total weight from 1 to 94 kg, covering a large range of THg BMI, from 0.008 to 0.35 (Table 1). The majority of shark species were juveniles or sub-adults, with a minimum pre-caudal length (PCL) of 56 cm for a silky shark individual, *Carcharhinus falciformis* (Table 1). Sexual maturity (adults) is around 2 m total length for *P. glauca, C. longimanus, C. falciformis* and *S. lewini*, and 3 m for *A. Superciliosus* (Drew et al., 2015; Chen et al., 1997; D'Alberto et al., 2017; Lessa et al., 2004; Grant et al., 2018). The highest mean THg concentration was found in *Prionace glauca*, which also presented the highest BMI (Table 1), indicating that the size (and the age) have a large influence on THg levels.

Positive and significant correlations were observed between THg concentrations and PCL for almost all species, except for *Prionace glauca* and *Carcharhinus longimanus* (Fig. 1B). No difference was found regarding the slopes of these correlations (ANCOVA, $p > 0.05$), traducing similar accumulation rates between species. However, when THg were normalized with the animal body mass (SBM), the highest values were observed in *Prionace glauca* and *Carcharhinus falciformis* ($R^2 = 0.35$ and 0.32 respectively). Maximum THg concentrations of our study were higher than those reported for all previous studies of these sharks (Table SI_2), except for *Carcharhinus falciformis* (O'Bryhim et al., 2017) and *Alopias pelagicus* (García-Hernández et al., 2007).

### 3.2. Mercury stable isotopes as indicators of sharks' Hg exposure

The Hg stable isotope fractionations are increasingly analyzed to explore the Hg biogeochemical cycle in marine environment, and more particularly the MIF used as a powerful tool to trace Hg sources. Our main focus is understanding Hg dynamics in top predator fish species, in the Eastern Equatorial Pacific, participating to a better knowledge of the mechanisms controlling Hg isotope variations in the pelagic Ocean.

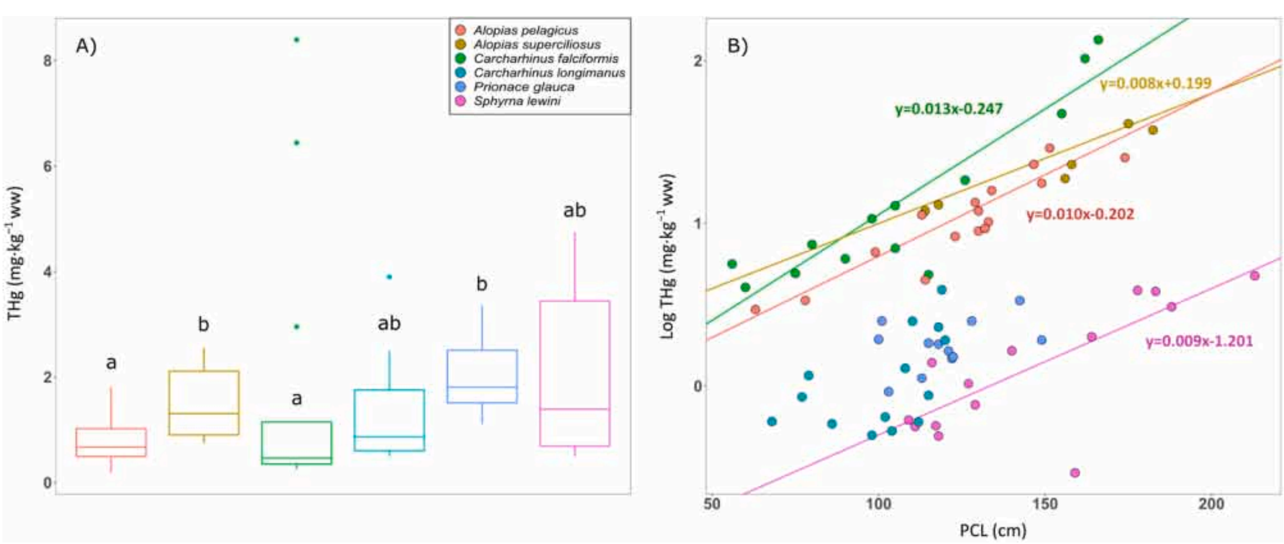


**Fig. 1.** A) Boxplots of total Hg concentration (THg, in µg g$^{-1}$ d.w.) in muscle of six shark species from the Eastern Equatorial Pacific. The box length represents the interquartile range, the bar length represents the range, and the horizontal line is the median value. Different letters (e.g. "a" and "b") indicate statistically significant differences between species ($p < 0.05$). B) Total mercury concentration (THg, in µg g$^{-1}$ d.w.) in log scale vs. pre-caudal length (PCL, in cm) for the same shark species. Data fit a linear regression for *A superciliosus, A. pelagicus, C. falciformis* and *S. lewini* ($p < 0.05$).

Under the influence of solar radiation, dissolved methylmercury can be transformed into inorganic Hg (iHg) by photodemethylation, while dissolved iHg can be converted into gaseous mercury Hg(0) by photoreduction. In aquatic experiments, MMHg photodemethylation is characterized by a $\Delta^{199}$Hg/$\Delta^{201}$Hg ratio of 1.36, while photodegradation of iHg leads to a ratio of 1.0 (Bergquist and Blum, 2007), these differences being explained by magnetic isotope effects (Buchachenko et al., 2007). In our study, the obtained slope of 1.20 between $\Delta^{199}$Hg and $\Delta^{201}$Hg (Fig. 2A) is explained by the dominance of MMHg demethylation over iHg photoreduction. This result shows that this MIF is caused by a difference of the nuclear volume effect, since MIF is produced by the influence of a difference in the nuclear spin with a slope ranging from 1 to 1.36 and by the effect of the magnetic nuclear champ with a slope ranging from 2 to 2.7 (Laffont, 2009). In addition, Hg photochemical degradation induces a $\Delta^{199}$Hg/$\delta^{202}$Hg slope of 2.4, whereas microbial transformation (no MIF) is characterized by a slope of 0 (Bergquist and Blum, 2007). In our study, the Hg isotopes in all shark samples displayed a $\Delta^{199}$Hg/$\delta^{202}$Hg slope of 1.54 (Fig. 2B), indicating the dominance of photochemical over microbial degradation. Our observations are similar to those previously reported in pelagic ecosystems of the Pacific Ocean (Blum et al., 2013; Madigan et al., 2018).

### 3.3. Mercury stable isotopes as indicators of sharks' foraging depth

Using $\Delta$ $^{199}$Hg and $\delta^{202}$Hg, it is possible to estimate the foraging depth of sharks (Le Croizier et al., 2020). Highest MIF and MDF values are usually related to surface feeding sharks, while sharks that feed in deep waters show lower $\Delta^{199}$Hg and $\delta^{202}$Hg. This variation is a consequence of the photochemical degradation of MMHg in sunlight surface water, resulting in elevated $\Delta^{199}$Hg and $\delta^{202}$Hg values (Blum et al., 2013). We found highest $\delta^{202}$Hg and $\Delta^{199}$Hg values in *Carcharhinus longimanus* (1.08‰ and 2.86‰ respectively) and *Carcharhinus falciformis* (0.96‰ and 2.89‰ respectively) (Table 1, Fig. 3), which is consistent with the known distribution of these species in the epipelagic zone (i.e. upper 200 m of the water column) (Andrzejaczek et al., 2018; Hutchinson et al., 2019). While the other shark species were characterized by lower $\Delta^{199}$Hg and $\delta^{202}$Hg values (Fig. 3), suggesting a foraging behavior associated with the mesopelagic zone (i.e. between 200 and 1000 m depth) (Bizzarro et al., 2017; Braun et al., 2019).

Indeed the two species of thresher sharks (*Alopias pelagicus* and *A. superciliosus*) are found from 0 to 700 m depth (Bizzarro et al., 2017). In the Pacific Ocean, *A. superciliosus* displays diel vertical migration and mainly occupies surface waters during the night, while it dives to 400–500 m during the day (Weng and Block, 2004; Nakano et al., 2003). The vertical distribution of *A. pelagicus* has received less attention, but Hg isotopes suggest a similar foraging depth to *A. superciliosus*, traduced by similar $\Delta^{199}$Hg and $\delta^{202}$Hg values between these two thresher shark species (Fig. 3A and B).

Most samples of *Prionace glauca* also showed low $\Delta^{199}$Hg and $\delta^{202}$Hg values (Fig. 3), which is congruent with deep feeding patterns of this species (Braun et al., 2019). Páez-Rosas et al. (2018) mention that *Prionace glauca* has a great diving capacity, allowing it to explore various habitats along the water column. In fact, it is capable of undertaking daily vertical migrations to depths of over 600 m in order to feed on mesopelagic cephalopods (Carey et al., 1990; Kubodera et al., 2007). Hg isotope signature show that *Sphyrna lewini* likely feed at the same depth as *P. glauca* (similar $\Delta^{199}$Hg and $\delta^{202}$Hg values; Fig. 3). Interestingly, *S. lewini* primarily occupies shallow waters (<100 m) near the Galapagos Islands, while it dives up to 300 m during offshore movements (Ketchum et al., 2014; Hearn et al., 2010). Here, Hg isotopes suggest that *S. lewini* may forage in the twilight zone when moving offshore, leading to low $\Delta^{199}$Hg $\delta^{202}$Hg values, comparable to those of thresher and blue sharks (Fig. 3).

Foraging depth generally increases during ontogeny in various shark species (Afonso and Hazin, 2015; Hoyos-Padilla et al., 2016), which would result in a decrease in $\Delta^{199}$Hg and $\delta^{202}$ Hg values with size. Interestingly, we found positive correlations between Hg isotopes and body length for 4 of the 6 species analyzed here: *P. glauca*, *S. lewini*, *A. pelagicus* and *A. superciliosus* (Fig. 4A and B). These trends could denote different foraging strategies at the intraspecific level, with larger individuals feeding at shallower depth. Although information is scarce on the reproductive cycle of these sharks, some species such as *S. lewini* are known to migrate between oceanic islands and pupping grounds in coastal areas (Salinas-de-León et al., 2017). Thus, the observed increase in $\Delta^{199}$Hg and $\delta^{202}$ Hg values with length could be due to a coastal signature of mature individuals. Further analysis covering the entire size range of these species would allow this hypothesis to be tested robustly.

In the North Pacific Ocean, Hg exposure has been shown to increase with foraging depth in pelagic fish (Choy et al., 2009; Madigan et al., 2018). In our shark samples, no correlation was found between Δ199Hg

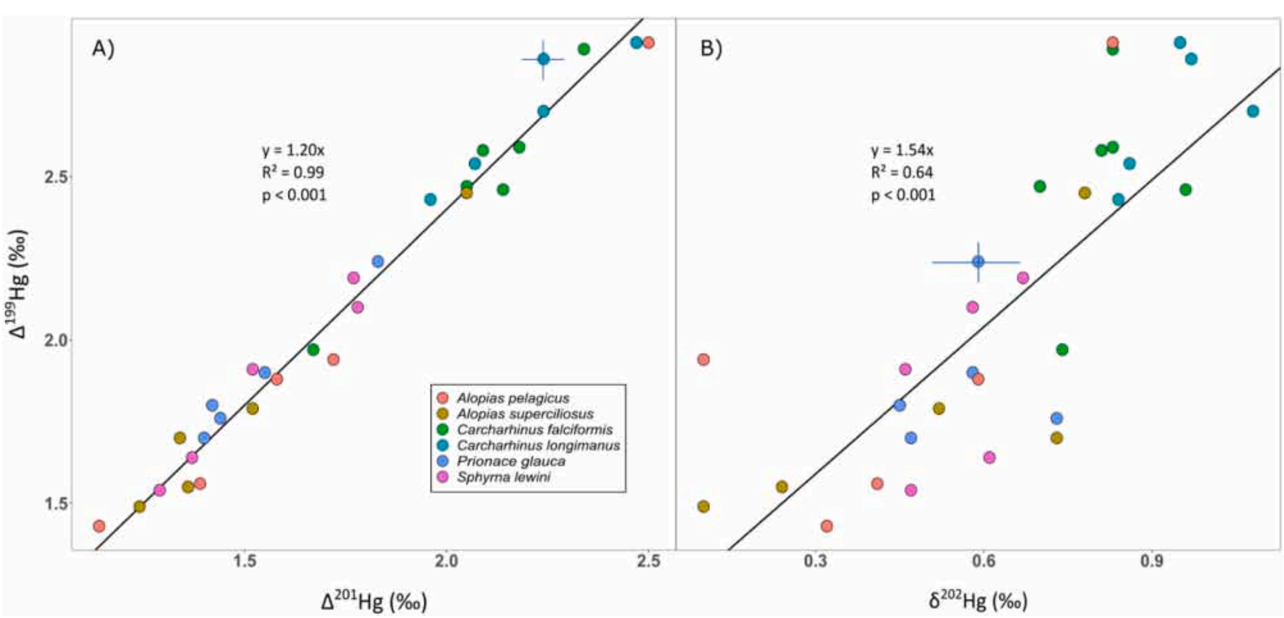


**Fig. 2.** Tri-isotopic ratios ( ± SD) A) $\Delta^{199}$Hg vs. $\Delta^{201}$ Hg and B) $\Delta^{199}$Hg vs. $\delta^{202}$ measured for the shark species samples in the Eastern Equatorial Pacific. Data fit a linear regression ($p < 0.001$).

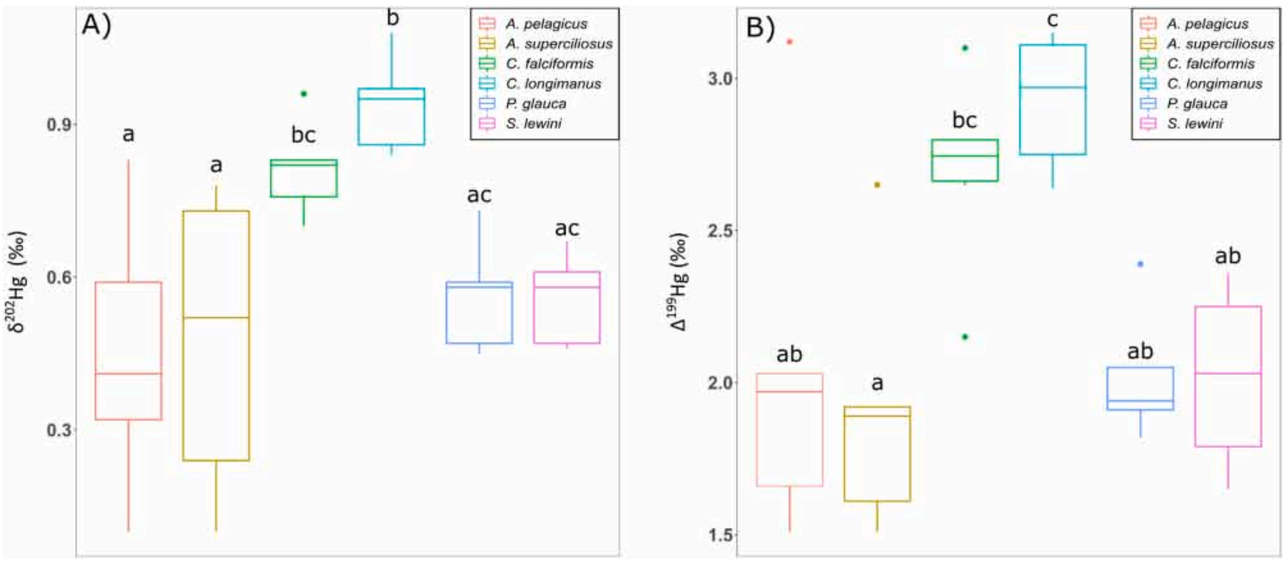


**Fig. 3.** Boxplots of Hg isotope signature (A: $\delta^{202}$Hg and B: $\Delta^{199}$Hg, in ‰) in muscle of six shark species from the Eastern Equatorial Pacific. The box length represents the interquartile range, the bar length represents the range, and the horizontal line is the median value. Different letters (e.g. "a" and "b") indicate statistically significant differences between species ($p < 0.05$).

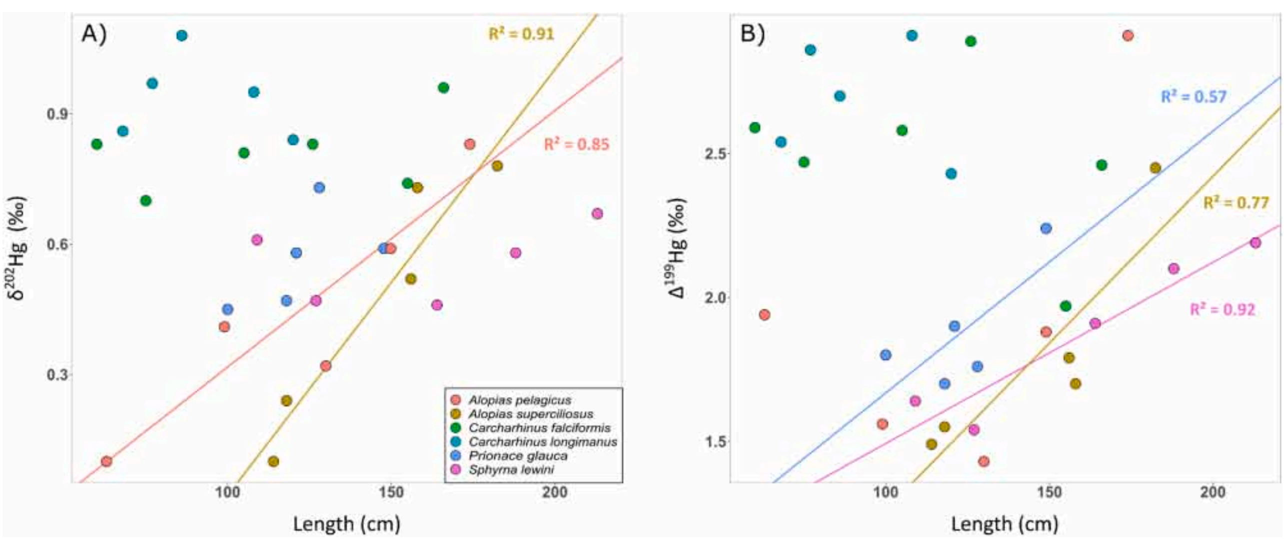


**Fig. 4.** A) $\delta^{202}$ Hg ($\pm$ SD) and B) $\Delta^{199}$Hg ($\pm$ SD) and vs. pre-caudal length (PCL) for shark species samples from the Eastern Equatorial Pacific. Significant linear regressions are shown ($p < 0.05$).

or δ202Hg and THg concentration (Figs. SI_2 and SI_3), suggesting that there is no influence of foraging depth on Hg exposure for sharks in the Equatorial Pacific. This result could, however, be masked by the predominant impact of other parameters such as size or trophic level on Hg concentrations.

In a previous study using carbon and nitrogen stable isotopes, significant overlap was found in the trophic niche of blue (*P. glauca*), silky (*C. falciformis*) and thresher (*A. pelagicus*) sharks from the Galapagos Islands (Páez-Rosas et al., 2018), suggesting competition for dietary resources between these species. Here, silky sharks displayed higher $\Delta^{199}$Hg and $\delta^{202}$Hg values than blue and thresher sharks (Fig. 3A and B), which means that strong trophic competition between these species is unlikely. Our study thus indicates that considering the vertical foraging habitat is essential for accurately assess resource partitioning in pelagic predators.

### 3.4. Human health risk assessment

In numerous cultures, women, men and children can be exposed to mercury contamination by regular sea-food ingestion and more specifically by shark ingestion all over the world (Vannuccini, 1999). Investigations in Asian countries [e.g., Cambodia (Agusa et al., 2007), Taiwan (Hsu et al., 2007), Japan (Sakamoto et al., 2007)] have reported fish/shellfish consumption levels greater than average worldwide consumption. Even in the US, Asian populations have higher MeHg intake than the Non-Asian population (Buchanan et al., 2015), seafood intake being a key predictor of blood Hg concentration (Liu et al., 2018). The consumption of shark species is a big concern, especially for its effects on fetus, newborn and children, as this element can cross the placenta barrier (Stern and Smith, 2003; Morrissette et al., 2004). In a less extent, neonates can also be exposed by consumption of contaminated breast-milk (Dorea and Barbosa, 2003). Thus, pregnant women and young mothers should be aware of THg exposure via predator marine fishes in their regular diet (World Health Organization, 2018).

Shark samples in this study show mean concentrations ranging from 0.77 μg THg $kg^{-1}$ up to 1.85 μg THg $kg^{-1}$ w.w., all exceeding the WHO reference value of 0.46 μg MMHg $kg^{-1}$. It is generally assumed that MMHg represents more than 90% of THg in the majority of shark species (Pethybridge et al., 2010; de Carvalho et al., 2014), but this is mainly based on teleosts while a recent study showed that mean MMHg percentage relative to THg in fins of eight shark species imported to mainland China and Hing Kong, was 69.0 ± 33.5% (Garcia Barcia et al., 2020). Silky shark was the species with a higher percentage of MMHg 82.10 ± 30.73% while scalloped hammerhead shark had the lowest percentage with 43.1 ± 24.2%. According to the US EPA's Guidance for Assessing Chemical Contaminant Data in Fish Advisories (2001), all the shark samples exceeded the threshold level of 0.12 mg MMHg $kg^{-1}$ w.w., defined for a consumption limit of 4 fish meals per month, and half of them (48%) for a consumption limit of 1 fish meal every two months. Table 2.

According to the World Health Organization (2018), the PTWI is 1.6 μg MMHg $kg^{-1}$ HBW. The tolerable weekly intake of THg by shark consumption is presented in Table 3 and depends on the human body weight and the shark species. By considering a median portion of 100 g of shark fillet in a single serving for an adult, we can observe that none of the analyzed species is edible, except for *A. pelagicus* (Table 3), limited at one portion per week. Thus, it is highly recommended to avoid the regular consumption of shark meat, more specifically for populations at risk such as pregnant women and young kids. Japanese authorities advise a maximum intake of shark muscle of 60–80 g per serving size, once a week or less for pregnant women (Japan Ministry of Health Labour and Welfare, 2003) which is higher than our estimations (Table 3). Considering the THg results in *S. Lewini, P. Lauca, C. longimanus, C. falciformis and A. Superciliosus*, the tolerable weekly amount of shark muscles ingested by men and women should not exceed 88 g and 75 g, respectively. While USA recommends avoiding the consumption of shark meat for pregnant women (Han and Watanabe, 2012).

Therefore, we can infer that the consumption of this particular meat may represent a serious human health risk. Nevertheless, shark meat represents a valuable source of proteins (Gordievskaya, 1973). This meat is highly appreciated because of its low fat contain and its high quantities of lysine, an important amino acid contained in fish meal (Kreuzer and Ahmed, 1978). Fish are also considered as an important source of n-3 fatty acids known for their protective health effects reducing risk of cardiovascular disease, stroke and diabetes; but these protective effects may be decreased by high levels of Hg as observed in tuna and sharks (Smith and Guentzel, 2010). In Taiwan shark consumption tends to be more popular among men than women because it is considered aphrodisiac (Fabinyi, 2012) and shark fin soup is commonly believed to have positive health benefits ranging from increasing virility to extending lifespan (Vannuccini, 1999). The MMHg concentration in dried, unprocessed fins are lower than in muscles, reducing the human Hg exposure. From a study realized in US restaurants, the average concentration of MMHg in shark fins reached 4.6 ng $mL^{-1}$ (n = 50); the consumption of a 240 mL bowl of shark fin soup containing would result in a dose of 1.1 μg MMHg, which is 16% of the U.S. EPA's reference dose for a 74 kg person; but, the soup containing the highest measured MMHg concentration (as measured in hammerhead sharks) would exceed the reference dose by 17% (Nalluri et al., 2014). More recently, in the Hong Kong markets, dried shark fins from five species surpassed methyl-mercury PTWIs (Garcia Barcia et al., 2020).

**Table 2**
Regression analysis of Log (THg) level and Pre-Caudal Length. Significant linear functions (Y=a+bx) were fitted. Y=Log (THg) level (mg $kg^{-1}$ ww), x is Pre-caudal Length (cm). The estimated intercept (a), slope (b), $R^2$, p value and number of samples (n) are listed by species.

| Common name | *Scientific Name* | a | b | $R^2$ | n | p value |
|---|---|---|---|---|---|---|
| Scalloped hammerhead | *Sphyrna lewini* | 0009 | 0003 | 0,59 | 14 | 0,0012 |
| Blue shark | *Prionace glauca* | 0004 | 0957 | 0,17 | 12 | 0,1837 |
| Oceanic whitetip | *Carcharhinus longimanus* | 0009 | 0317 | 0,28 | 14 | 0,0525 |
| Silky shark | *Carcharhinus falciformis* | 0013 | −0247 | 0,82 | 13 | 0,0000 |
| Bigeye thresher shark | *Alopias superciliosus* | 0008 | 0199 | 0,92 | 6 | 0,0025 |
| Pelagic thresher | *Alopias pelagicus* | 0010 | −0202 | 0,84 | 14 | 0,0000 |

**Table 3**
THg measured average concentration per study shark species and tolerable weekly intake of shark meat (in g) for men, women and children.

| Shark specie | THg average concentration (mg $kg^{-1}$ ww) | Average body weight (kg) Men 70 | Women 60 | Children 15 |
|---|---|---|---|---|
| | | Weekly intake (g shark/weekly) | | |
| *Sphyrna lewini* | 1.75 | 64 | 55 | 14 |
| *Prionace glauca* | 1.85 | 61 | 52 | 13 |
| *Carcharhinus longimanus* | 1.28 | 88 | 75 | 19 |
| *Carcharhinus falciformis* | 1.74 | 64 | 55 | 14 |
| *Alopias superciliosus* | 1.49 | 75 | 64 | 16 |
| *Alopias pelagicus* | 0.77 | 145 | 125 | 31 |

*PTWI (μg MeHg $kg^{-1}$ HBW):1.6

Considering the human health risk assessment results, the consumption of shark meat represents a serious human health risk for the populations that include shark in their regular diet. Special attention should be focused on pregnant women and children concerning the frequency and amount of shark meat intake.

## 4. Conclusions and recommendations

The concentration of THg and, therefore, of MMHg in sharks is highly influenced by their body size (age) and dietary habits. Our data show that none of the THg levels of the shark species seized from the 'Fu Yuan Yu Leng 999' in the Galapagos Reserve is tolerable for a weekly consumption; thus, shark meat represents a serious human health risk for the populations that include these predator fishes in their regular diet. The study of Hg isotopes helps to explain the biology and feeding patterns of large pelagic fish species. Most of shark species have slow growth rates, late age-at-maturity and low fecundity compared with bony fishes. These life history parameters result in a limited ability to withstand fishing pressure and a longer recovery time in response to overfishing. The fishing activities are illegal in the Galapagos Marine Reserve, known as the largest global shark biomass of the world (Salinas de León et al., 2016). Sustainable fisheries for sharks are possible, but need to be closely managed with small yields compared to standing stocks (FAO, 2019). The increase in effort and the expansion of the areas fished in the recent decades has led to concerns over the consequences for the populations of several shark species in the world's oceans, and particularly in the Eastern Equatorial Pacific. Many exploited species of sharks are declining and several have been protected by national legislations and international treaties, like the CITES (Convention on International Trade in Endangered Species of wild Fauna and Flora), which aims to ensure that international trade in specimens of wild animals and plants does not threaten their survival. However, the estimation of the impacts of CITES on shark's listing in Southeast Asian fisheries (Friedman et al., 2018) is biased by the lack of data from Asian countries production and captures.

## CRediT authorship contribution statement

**Laurence Maurice**: Conceptualization, Methodology, Formal analysis, Supervision, Writing - original draft, Writing - review & editing. **Gaël Le Croizier**: Formal analysis (Hg stable isotopes), Statistical treatment, Writing - review & editing. **Gabriela Morales**: Formal analysis (THg), Writing - review & editing. **Natalia Carpintero**: Formal analysis (THg). **Juan M. Guayasamin**: Formal analysis (genetic species determination), Logistics, Writing - review & editing, Funding acquisition. **Jeroen Sonke**: Formal analysis (Hg isotopes Analysis supervision), Writing - review & editing. **Diego Páez-Rosas:** Research Permits**,** Logistics, Writing - review & editing. **Walter Bustos**: Research Permits**,** Logistics. **David Point**: Writing - review & editing, Funding acquisition. **Valeria Ochoa-Herrera**: Formal analysis (THg), Writing - review & editing, 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. Supporting information

Supplementary data associated with this article can be found in the online version at doi:10.1016/j.ecoenv.2021.112122.

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