Reviews of Environmental Contamination and Toxicology (2022) 260:4
https://doi.org/10.1007/s44169-021-00006-2

REVIEW

A Global Review of Cadmium, Mercury, and Selenium in Sharks: Geographical Patterns, Baseline Levels and Human Health Implications

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Received: 9 July 2021 / Accepted: 21 October 2021

Abstract
Globally, there is concern about the concentrations of some metals and metalloids in sharks and the associated impacts to themselves and to human consumers. Concentrations of Hg, Cd, and Se reported in muscle, liver and fins of sharks from all the world were reviewed to define geographical patterns of elemental distribution, to model the baseline concentrations of the three elements, and to assess potential human health risk. Published information corresponded to 102 sites that belong to thirteen FAO Major Fishing Areas. The majority of the sites corresponded to the Atlantic (39 sites), followed by the Pacific Ocean (37). Statistical analysis indicated significant differences of elemental concentrations among all FAO regions and oceans. The 5% baseline levels, estimated with a cumulative frequency distribution method for each element were: for Hg, 0.129 µg g^{-1} in muscle, 0.147 µg g^{-1} in liver; for Cd, 0.517 µg g^{-1} in muscle, 0.290 µg g^{-1} in liver; for Se, 0.105 µg g^{-1} in muscle, 0.218 µg g^{-1} in liver. These baseline leves are equivalent to the 5th percentile on a normal distribution. Levels of Cd, Hg and Se in muscle of sharks were above maximum permissible limits (Cd, 0.5; Hg, 1.0; Se, 1.0 µg g^{-1}; Nauen in FAO Fish Circ 764:1–102, 1983; CEC in Off J Eur Union 364:5–24, 2006) for human consumption in 6, 49 and 25% of the reports, respectively. Considering all the studies, hazard quotients (it indicates the risk during the lifetime of an individual different to cancer if value is above one) were Cd (0.10 ± 0.18), Se (0.11 ± 0.10) and Hg (2.05 ± 2.69); i.e. only average Hg values are of concern. The concentrations of Hg and Se in muscle of sharks from the Mediterranean were statistically different from all other oceans. In shark species with more reports, baseline levels (µg g^{-1} wet weight) of Hg in muscle were: for *P. glauca* from the Pacific Southeast (0.048), for *I. oxyrinchus* from the Pacific Southeast (0.034), and for *S. lewini* from the Pacific Eastern Central (0.55). The balance of Hg and Se molar concentrations (it is used to assess the benefit or risk to the combined occurrence of Hg and Se) in muscle of sharks was assessed through the Se health benefit value (HBV_{Se}), positive results indicate that shark consumption is beneficial to humans. In general, HBV_{Se} was positive (3.06 ± 13.84). According to FAO Fishing areas, negative HBV_{Se} values corresponded to Mediterranean and Black Sea (− 48.37), Atlantic Northwest (− 5.54), Pacific Eastern Central (− 0.97), and Atlantic Southwest (− 0.21); nevertheless, HBV_{Se} values in sharks from Mediterranean and Black Sea, and Atlantic Northwest corresponded to only one species. Overall, similar elemental concentrations were found in some FAO regions of the southern hemisphere. Baseline values of Hg were in the same magnitude order as the low atmospheric deposition flux. Under the approach of the hazard quotient, Hg is of concern because values were above one in almost half of the studies. More information related to Hg and Se in muscle of sharks is required in some FAO regions (Indian Ocean Eastern, Pacific Northeast and Pacific Southeast) to estimate HBV_{Se}.

Introduction

Diverse elements are necessary for the correct functioning of aquatic biota (Perelló et al. 2008), such elements are known as essential and their occurrence is preferable within certain limits depending on the organism (metabolism, species, feeding habit, age/size, etc.) and the element (chemical species and bioavailability). Although essential elements are necessary for metabolism, at elevated concentrations they may

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Published online: 22 January 2022

produce deleterious effects. Selenium (Se) is an essential element that occurs naturally in the environment but it is also supplied through anthropogenic activities (Páez-Osuna and Osuna-Martínez 2015), mainly as a by-product of agriculture, coal extraction and mining of copper, uranium and phosphate (Young et al. 2010). Since Se is supplied from natural and anthropogenic sources, it may be found globally. There is another group of elements termed as non-essential, which are not necessary for metabolism and at low concentrations may cause lethal and sublethal effects. Cadmium (Cd) and mercury (Hg) are non-essential elements and they are supplied to the environment by natural processes and human activities. On a global scale, the main contribution of Cd to the atmosphere (Pacyna and Pacyna 2001) is due to the production of non-ferrous metals (73%), and the combustion of fossil fuels (23%) with a flux to the atmosphere from natural and anthropogenic sources of 1300 t y^{-1} and 7600 t y^{-1}, respectively (Millward and Turner 2010). Conversely, volatile Hg and organomercury compounds are emitted from the land into the atmosphere naturally from wildfires, volcanoes, and microbial activity. Anthropogenically, Hg is emitted into the atmosphere as a result of high temperature processes (e.g., smelting, coal combustion, incineration) combined with commercial uses of elemental Hg (e.g., batteries, thermometers), as well as disposal of Hg-laden wastes from gold mining operations that are converted into volatile forms in the environment. Natural and anthropogenic Hg emissions from the land into the atmosphere are estimated to be 2500 and 3600 t y^{-1}, respectively (Millward and Turner 2010).

Essential and non-essential elements are accumulated by aquatic fauna in varying levels, depending on the element, the organism, and the degree of impact of the water body. In the case of fish, it is known that species of elevated trophic levels may accumulate higher concentrations of pollutants than specimens of low positions in the food web (Ali and Khan 2018). The habitat of sharks is also an important factor in the context of pollutant accumulation since potential preys of the diverse habitats (demersal, benthopelagic, pelagic, etc.) may vary in their content of essential and non-essential elements. Sharks are usually top predators that may accumulate elevated levels of diverse elements; in fact, marine ichthyofauna are considered as some of the main sources of Cd (Okocha and Adedeji 2011), Se (Navarro-Alarcon and Cabrera-Vique 2008) and Hg (Fréry et al. 2001) to humans. Considering that sharks are distributed worldwide, they can be used as tracers of diverse pollutants. In fact, there is a high concern associated to the elevated levels of some trace metals in sharks (Alves et al. 2016) but the impact of pollutants on their health is not well understood (Tiktak et al. 2020). Though some species migrate considerable distances, geographical trends of elemental accumulation may be established at broad regional scales (*i.e.,* ocean basins). In this context, Pethybridge et al. (2010) found Hg differences in diverse shark species from Australia in comparison with other regions of the world; however, the authors did not report elemental differences at smaller geographical basis. Similarly, in a study with the Greenland shark *Somniosus microcephalus* and the Pacific sleepershark *Somniosus pacificus* from Arctic waters (McMeans et al. 2007), it was found that several non-essential elements (including Cd and Hg) were significantly different between sharks from the Atlantic and the Pacific Ocean. On the contrary, McMeans et al. (2007) found no differences of Se levels between *S. microcephalus* and *S. pacificus* from two distant areas. Perhaps it was due to the similar capacity of both shark species to regulate the levels of Se, combined with a comparable exposure to this metaloid. In the context of this paper, most of the studied sharks are commercially important pelagic species that spend most of their lifecycle in the open sea, and therefore our results should reflect oceanic patterns (Coiraton and Amezcua 2020; Rogers et al. 2015; Vandeperre et al. 2016).

Background concentrations of diverse elements denote their pre-industrial or natural levels from pristine areas (Lu et al. 2019). However, under the long-term influence of human activities, the original metal(oid) concentrations of worldwide natural environments have essentially become absent. The baseline concept is sometimes used in equivalency to ambient background in the context of measuring levels “now” such that future change can be quantified (Reimann and Garret 2005). Baseline levels could characterize any area or region evidencing the heterogeneity of the environment (Lu et al. 2019). Sharks, like other organisms, exhibit a metal(oid) bioaccumulation that is an integrated time-dependent process, which involve various strategies to accumulate metal(oid)s from a wide range of waters and food. Accordingly, metal(oid)s tissue concentrations may vary several orders of magnitude under distinct and changing geographic environments. Consequently, it is important to assess the baseline tissue metal(oid) levels in specific shark species or group of shark species.

Marine fish contribute with elevated amounts of certain elements to humans (Okocha and Adedeji 2011) so it is necessary to measure their concentrations and assess the risk associated to fish consumption. In the case of Cd and Hg, their levels might be biomagnified and constitute a health problem. Contrastingly, several nutrients and Se may provide diverse benefits, including protection against Hg toxicity (Ralston 2008). In the present study, concentrations of Hg, Cd, and Se reported in muscle, liver and fins of sharks from all over the world were reviewed to define geographical patterns of elemental distribution, and to assess potential human health risk. Simultaneously, we applied the cumulative frecuency distribution (CFD) method to model the baseline concentrations of the three elements.

Materials and Methods

Data Collection: Database

Published literature was found through two search engines, Google Scholar (https://scholar.google.com) and Scopus (https://www.scopus.com), using a combination of the following keywords: "sharks", "elasmobranchs", "muscle", "liver", "fins", "trace metals", "elemental concentrations", "selenium", "cadmium", and "mercury". The retrieved articles were then screened by region, species, tissues, and element. Only studies on Cd, Hg and Se in muscle, liver, and fins of any shark species (Selachimorpha) were selected.

The analytical quality of the generated data on the elements is crucial; consequently, a review of the methodologies was made. The results showed that 81.2% of the studies reported the use of reference materials and obtained reasonable recoveries (85–115%). The rest of the studies could have generated acceptable or unacceptable data; however, since its percentage was relatively low, it was decided to work with the total data. Elemental concentrations are given as $\mu g\ g^{-1}$ on wet weight basis; if original information was provided on dry weight basis, conversion was made assuming the humidity percentage for muscle (75%), liver (65%), and fins (70%) as reported in diverse studies (Kovekovdova and Simokon 2002, for liver; Man et al. 2014, for fins; Gil-Manrique et al. 2017, for muscle). To assess regional variations of elemental concentrations, thirteen FAO fishing areas (FAO 2015) were considered, namely, Pacific Northwest, Pacific Northeast, Pacific Western Central, Pacific Eastern Central, Pacific Southwest, Pacific Southeast, Atlantic Northwest, Atlantic Northeast, Atlantic Western Central, Atlantic Southwest, Mediterranean and Black Sea, Indian Ocean Western, and Indian Ocean Eastern.

Information Analysis and Statistics

A total of 102 sites (Fig. 1) were analyzed and grouped into thirteen regions according to the FAO Major Fishing Areas (Table 1). One-Way ANOVA was used to test for differences in mean concentrations of Hg, Se and Cd between liver and muscle. The assumption of homoscedasticity was tested with Cochran´s C test. The test was performed on STATISTICA

Table 1 Major FAO fishing areas and correspondent sites where studied sharks were collected and studied for Cd, Hg and Se concentrations in muscle

FAO fishing area	Site
Atlantic, Northwest	Cumberland Sound, Canada. Long Island, NY. New England, USA. New York Bight. NW Atlantic (Massachusetts, USA). NW Atlantic, USA
Atlantic, Northeast	Atlantic Ocean. Azores (Mid-Atlantic). Basque Country, Spain. Celtic Sea. Cornwall, England. English Channel. Irish sea. Liverpool Bay. NE Atlantic. NE Greenland. Rockall Trough. Southwest Portugal. Valencia, Spain. West coast of Ireland
Atlantic, Southwest	Bahía Blanca estuary, Argentina. Brazilian offshore waters (13°22' S). Buenos Aires, Argentina. Equator (1–5° S; 21–25° W). Rio de Janeiro, Brazil. Rio de la Plata, Argentina. SE Brazil. Southern Brazilian waters
Atlantic, Western Central	Atlantic Ocean (25°006' N; 81°042' W). East Florida. Florida. Georgia and Florida, USA. Gulf of Mexico and Caribbean. NE coast of USA (Georgia, SC, FL). Southern Florida. SW Florida. Tamiahua, Mexico. Trinidad and Tobago. Veracruz, Gulf of Mexico
Indian Ocean, Eastern	Bay of Bengal. Derwent estuary, Tasmania. Gulf of Andaman. Malaysia. Southern Australia. Tasmania
Indian Ocean, Western	East coast of South Africa. Eastern Madagascar. Gulf of Aden. Gulf of Oman. Indian ocean (10° S; 75° E). Kuwait. Langebaan lagoon, South Africa. Persian Gulf. South Africa coast. Southeastern Madagascar. SW Indian ocean
Mediterranean and Black Sea	Aegean Sea. East Mediterranean Sea (Crete). Eastern Mediterranean (32°31' N; 34°02' E). Ionian Sea. Marmara Sea, Turkey. Mediterranean Sea. Mediterranean Sea. NW Mediterranean Sea. SE Mediterranean sea (Italy). Tyrrhenian sea
Pacific, Eastern Central	Altata, SE Gulf of California. California. Costa Rica. East coast of the Gulf of California. Gulf of California. Hawaii. Isla Magdalena, BCS. Mazatlán, Mexico. North Pacific Ocean near Hawaii. San Francisco, USA. SE Gulf of California. Southern Baja California. Southern California. Tropical eastern Pacific (Mexico). West coast of Baja California Sur
Pacific, Northeast	NE Pacific, USA. Oregon coast. Strait of Georgia, British Columbia
Pacific, Northwest	Ishigaki Island, Japan. Chiba Prefecture, Japan. Hong Kong. Ishigaki island, Japan. Jeju Island, Republic of Korea. Korean coastal waters. Northern Japan
Pacific, Southeast	Chile (21–35° S; 78–118° W). Northern Peru
Pacific, Southwest	New South Wales, Australia. New Zealand. SE Australian waters. South Island, New Zealand
Pacific, Western Central	NE Australia (Queensland coast). New Guinea and Australia. Northern Territory, Australia. Papua, New Guinea. Queensland, Australia

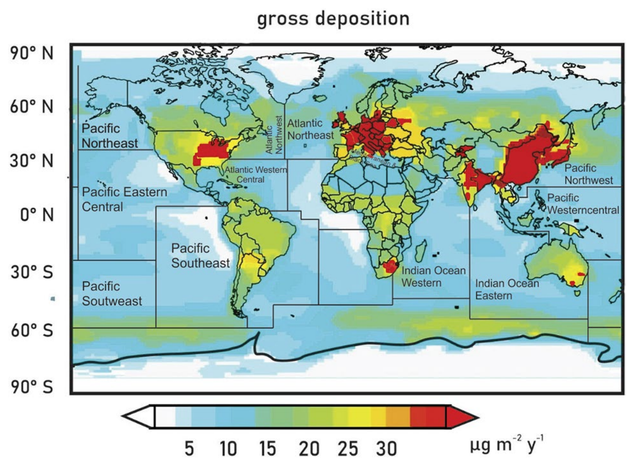

Fig. 1 FAO Fishing Areas where sharks were collected, and global atmospheric deposition of Hg (μg m^{-2} y^{-1}). Red shades indicate high (>30) atmospheric deposition, blue and white shades indicate low (<15) atmospheric deposition of Hg (Color figure online)

Version 12 (StatSoft). Permutational multivariate analysis of variance (PERMANOVA) was used to determine differences in the concentration of Hg, Cd and Se across all shark species according to the factors tissue (muscle and liver), trophic levels (obtained from FishBase), years, ocean and FAO regions. To do this, first the concentrations of all three elements were normalized using PRIMER, given that these data involve measurements with different scales. The normalization routine transforms every variable, such that they each has a mean of 0 and a standard deviation of 1. After normalized element concentrations/tissue data were 2nd root transformed, and a matrix containing every element in every tissue and every site per year as columns and shark species as rows was created, and the previously mentioned factors were assigned to each column. From this, a Euclidean distance similarity matrix was generated. Pairwise-tests were performed to determine which factors differed from the others, and a Bonferroni procedure was applied to correct for multiple comparisons. If significant results were found, the data were graphically represented using a Principal Coordinates test (PCO) (McArdle and Anderson 2001).

A similarity of percentages (SIMPER) analysis was used to determine which shark species accounted for most of the dissimilarities among the element concentrations in the different factors when significant differences were found (Collins and Williams 1982; Clarke and Warwick 1994), and thereby identified characteristic species for each site according to the relation Sq.Dist/SD (square distance/standard deviation). All multivariate statistical analyses were completed using the PRIMER 6 statistical package with the PERMANOVA + add-on (PRIMER-E, Plymouth Marine Laboratory, UK).

Baseline levels were estimated using the cumulative frecuency distribution method to model the baseline concentrations of the three elements in liver and muscle (Lu et al. 2019). In our study, we operationally defined the 5% distribution as the baseline metal(oid) concentration, which is consistent with most of the environmental quality assessments based on the frequency distribution of different environmental parameters (Lu et al. 2019). For Cd and Se, information on global atmospheric deposition is non-available, consequently, it was assumed to be proportionally similar to Hg. Atmospheric deposition of Hg contributes significantly to the surface ocean mixed layer, but the deep ocean also accounts for a substantial quantity of Hg inputs to the surface ocean mixed layer (Zhang et al. 2016). For the current study, information of Hg deposition from Zhang et al. (2016) was adapted to include FAO Fishing areas. Conversely, the limitation of this approach is the mobility of studied sharks; *i.e.,* they could cover areas with high Hg deposition fluxes. In this study, principal component analysis (PCA) was employed to reveal the clustering pattern that identifies sites with more anthropogenic inputs of elements. Baseline concentrations were calculated based on 95th percentile of the dataset (Parkman 2007; Lu et al. 2019). Application of multivariate approaches such as PCA for the assessment of the baseline values and identification of uncontaminated sites has been widely reported (Guan et al. 2016; Sim et al. 2016).

Human Health Risk Assessment

Comparison of elemental concentrations in muscle with maximum permissible limits were made using wet weight values; maximum permissible limits in fishery products were: Cd, 0.5 (Nauen 1983); Hg, 1.0 (CEC 2006), and Se 1.0 μg g^{-1} (Nauen 1983). Human health risk associated to the levels of the studied elements and consumption of shark was estimated by the hazard index ($HI = HQ_{Cd} + HQ_{Hg} + HQ_{Se}$); it indicates if there is a risk during the lifetime of an individual different to cancer according to the levels of Cd, Hg and Se in muscle of sharks and the rate of consumption of shark muscle. The HI was calculated as the addition of the hazard quotients (HQ) for Cd, Hg, and Se. For the estimation of HQ we used the concentrations of Cd, Hg, and Se in the muscle of shark using the equation HQ = E/RfD (Newman and Unger 2002). In the equation, E is the exposure to Cd, Hg, and Se through muscle shark consumption, and RfD is the reference dose (US EPA 2000) of the elements of interest (Cd, 0.5 μg kg^{-1} body weight day^{-1}; Hg, 0.3 μg kg^{-1} body weight day^{-1}; Se, 5 μg kg^{-1} body weight day^{-1}). The exposure level (E) is calculated as $E = C \times I/W$; C is the concentration of the element of interest in shark muscle, I is the average ingestion rate of shark (41.9 g day^{-1}) in the world (Laureti 1998) and W is the weight of an average adult (62 kg) according to Walpole et al. (2012). Additionally, in order to assess the benefit or risk due to the combined occurrence of Hg and Se in shark muscle, the HBV_{Se} was used following the approach of Ralston et al.

(2016). The HBV_{Se} = ([Se–Hg]/Se) x (Se + Hg) considers the concentrations of Se and Hg in μmol kg^{-1} in shark muscle; positive results indicate that shark consumption would improve Se status in humans, negative values mean the opposite. The magnitude of the HBV_{Se} value indicates Se surplus or deficit associated to shark consumption.

Results and Discussion

Shark Species: Geographical Distribution, Habitat, and Trophic Position

A total of 358 individual records from 124 publications (Supplementary material-references) were included, they contained shark information of 21 families, 46 genera and 113 species; the trophic position (TP) of sharks (Froese and Pauly 2021) ranged from 3.4 to 4.7 (mean 4.1 ± 0.3), although only one species had a TP below 4. The number of reports varied depending on the tissue and the element; muscle had more records (Cd, 100; Hg, 304; Se, 69) than liver (Cd, 65; Hg, 77; Se, 23) and fins (Cd, 5; Hg, 23, Se, 1). The collection time of sharks ranged from 1971 to 2018. The habitat of collected specimens were: demersal (27.4%), benthopelagic (22.3%), benthopelagic-reef (21.2%), pelagic (19.8%), bathydemersal (6.4%), and reef associated (2.8%). Most of the collected specimens included high migrant and circumtropical species such as thresher sharks (*Alopias* sp), requiem sharks (*Carcharhinus* sp), mako (*Isurus* sp), blue sharks (*Prionace* sp), hammerheads (*Sphyrna* sp), and the great white shark (*Carcharodon carcharias*) which is a cosmopolitan species. Other important non-migrant species were tiger sharks (*Galeocerdo* sp), houndsharks (*Mustelus* sp), and sharpnose sharks (*Rhizoprionodon* sp). Few studies focused on non-migrant and estuarine species, such as *Triakis* sp, and reef associated such as *Triaenodon* sp.

The majority of the studied sharks are commercially important and circumtropical species, therefore, studies on them have been undertaken in many regions. In fact, the most studied species was the blue shark (*Prionace glauca*, 22 studies), which is also the most common and the one with the largest global landings of all sharks in the FAO database (Musick and Musick 2011). Something similar happens with species from which there are more than 10 published works with the exception of the Blackmouth catshark (*Galeus melastomus*) which is very important in the landings from Europe and the Mediterranean, and the Picked dogfish (*Squalus acanthias*) of commercial importance in temperate and cold areas such as the north Atlantic.

It is necessary to point out that the existence of studies within a region depends on two factors, first of all, the abundance and diversity of sharks in the waters of each area, and second on the interest in carrying out the studies, which may be driven by the fisheries, the potential environmental impacts as well as funding and the presence of research institutions in the region.

Geographically, most of the studies have been undertaken in the Eastern Central Pacific, which corresponds to the north part of the Tropical eastern Pacific, Baja California in Mexico and most of California and Hawaii in the USA. In terms of regions, 99 studies corresponded to Oceania (Australia and New Zealand, FAO Areas Eastern Indian Ocean and Pacific Western Central and Southwest), followed by north America (Canada, USA and Mexico, FAO areas Pacific Eastern Central, Atlantic Western Central, Pacific Northeast and Atlantic northwest) where 97 studies were undertaken. In the third place is Europe (FAO area Atlantic Northeast and Mediterranean), where 61 studies were found. These studies account for approximately 72% of all the studies analyzed in this paper.

Cd, Hg, and Se According to Tissue, Geographical Area, and Habitat

Considering all sharks, ANOVA revealed statistical differences in the mean concentrations of Hg and Se between muscle and liver (Hg $F_{(1, 392)} = 9.4729$, $p < 0.01$; Se $F_{(1, 89)} = 22.661$, $p < 0.01$), but no differences were found in the mean concentrations of Cd ($F_{(1, 162)} = 3.6603$, $p > 0.05$) between these tissues (Table 2). The sequence of elemental concentrations in muscle was Hg (1.507 μg g^{-1}), followed by Se (0.785 μg g^{-1}) and Cd (0.153 μg g^{-1}). In the liver, the order was Cd (7.963 μg g^{-1}), Se (3.88 μg g^{-1}) and Hg (1.566 μg g^{-1}). In the case of fins, the sequence of elemental concentrations was Hg (0.828 μg g^{-1}), Cd (0.745 μg g^{-1}) and Se (0.35 μg g^{-1}). The shark species with the highest number of reports of elemental concentrations were *P. glauca* ($n = 22$), *I. oxyrinchus* ($n = 21$) and *S. lewini* ($n = 15$). In the three species, muscle was the tissue with the most elevated concentrations of Hg. It is noticeable the elevated variability of elemental concentrations in all tissues; it was expected to find contrasting concentration of all elements since calculations include individuals of varying ages, trophic levels and feeding habits. With respect to the average concentrations of analyzed elements in the studied tissues, liver had more elevated levels of Cd, Hg and Se than muscle and fins; such behavior is similar to the sequence of Hg levels in sharks from Mexican waters (e.g., Barrera-García et al. 2012; Bergés-Tiznado et al. 2015; Páez-Osuna et al. 2017; Ruelas-Inzunza et al. 2019).

PERMANOVA revealed no statistically significant differences according to the TP (pseudo-$F_{2,83} = 1.1212$, $p = 0.283$), and year of collection (pseudo-$F_{5,83} = 0.7732$, $p = 0.646$). However, significant discrimination among FAO region and Ocean were observed (FAO Region: pseudo-$F_{13,70} = 2.082$, $p = 0.041$; Ocean: pseudo-$F_{4,79} = 1.599$, $p = 0.05$). FAO regions from the same ocean grouped close to each other,

Table 2 Summary of elemental concentrations (μg g^{-1} wet weight) in selected tissues of sharks from all FAO regions

Species	Tissue	Parameter	Cd	Hg	Se
Prionace glauca	Muscle	Mean (± SD)	0.19 (0.18)	0.90 (0.59)	0.25 (0.12)
		N	5	16	8
		Median	0.2	0.89	0.26
		Range	0.01–0.45	0.048–2.25	0–1 to 0.45
	Liver	Mean (± SD)	10.1 (16.5)	0.54 (0.44)	1.77 (0.12)
		N	4	4	3
		Median	2.71	0.39	1.74
		Range	0.25–34.66	0.22–1.17	1.67–1.91
	Fins	Mean (± SD)	–	2.4	–
		N	–	1	–
		Median	–	2.4	–
		Range	–	–	–
Isurus oxyrinchus	Muscle	Mean (± SD)	0.50 (0.59)	1.33 (1.1)	0.38 (0.13)
		N	6	17	5
		Median	0.29	1.16	0.36
		Range	0.00005–1.32	0.206–3.36	0.237–0.6
	Liver	Mean (± SD)	0.85 (0.79)	1.1 (1.5)	–
		N	2	2	–
		Median	0.85	1.1	–
		Range	0.29–1.41	0.0000001–2.2	–
	Fins	Mean (± SD)	–	3.2	–
		N	–	1	–
		Median	–	3.2	–
		Range	–	–	–
Sphyrna lewini	Muscle	Mean (± SD)	0.02 (0.01)	1.09 (0.66)	0.86 (0.38)
		N	5	13	4
		Median	0.02	0.82	0.95
		Range	0.0012–0.025	0.55–3.11	0.34–1.095
	Liver	Mean (± SD)	0.10 (0.09)	0.14 (0.07)	5.9 (2.42)
		N	2	5	2
		Median	0.10	0.14	5.9
		Range	0.035–0.168	0.042–0.23	4.27–7.7
	Fins	Mean (± SD)	–	–	–
		N	–	–	–
		Median	–	–	–
		Range	–	–	–
All	Muscle	Mean (± SD)	0.15 (0.62)	1.51* (1.979)	0.79*(0.735)
		N	81	301	69
		Median	0.043	0.99	0.59
		Range	1.32–0.00005	18.29–0.007	3.32–0.05
	Liver	Mean (± SD)	7.96 (38.033)	1.57*(1.823)	3.90* (5.310)
		N	56	75	22
		Median	0.916	1.11	1.703
		Range	284.5–0.00013	12.93–0.01	24.7–0.41
	Fins	Mean (± SD)	0.75(0.792)	0.83 (0.865)	0.35
		N	9	21	1
		Median	0.514	0.514	0.35
		Range	2.4–0.091	3.2–0.007	0.35–0.35

*For a given column, concentrations were statistically different but most of these regions were statistically different. Pair-wise comparisons of the PERMANOVA testing for differences between FAO regions (Table S1) and oceans (Table S2) indicate that the studies from which no information was given regarding the location were mixed between all oceans, and the concentrations in these areas were statistically equivalent. The Mediterranean showed element values which were separated clearly to the right of the figure and the concentration of Hg and Se in the muscle was statistically different from all the other oceans (Fig. 2). The Indian Ocean formed a group to the left of the graph along the vertical axis, with the concentrations being different from the Mediterranean and Atlantic, but being statistically equivalent to the Pacific. In the Indian Ocean some parts showed higher concentrations of Se in liver and Cd in muscle (bottom), while other showed a higher concentration of Hg and Cd in liver. Atlantic and Pacific Oceans, formed defined groups, most of the Pacific regions group together in the bottom right part of the figure, and most of the Atlantic regions grouped in the bottom left part. In the Pacific in general the concentration of Hg in both muscle and liver and Se in muscle are higher, and in the Atlantic Ocean the concentration of Cd in both liver and muscle are higher (Fig. 2).

In terms of element and tissue, PERMANOVA showed statistical differences between the concentrations of the different metals across both tissues (pseudo-$F_{5,83} = 1.24$, $p = 0.05$). When looking into the pair-wise comparisons, the concentration of Hg in muscle was different to all the other combinations, and the concentrations of Hg in liver, and Cd and Se in muscle and liver were not different among themselves (Table S3). From results in SIMPER (Table S4), it is evident that the concentration of Hg in muscle was significantly higher than the other concentrations, even Hg in liver. The sharks that most contributed to these differences, according

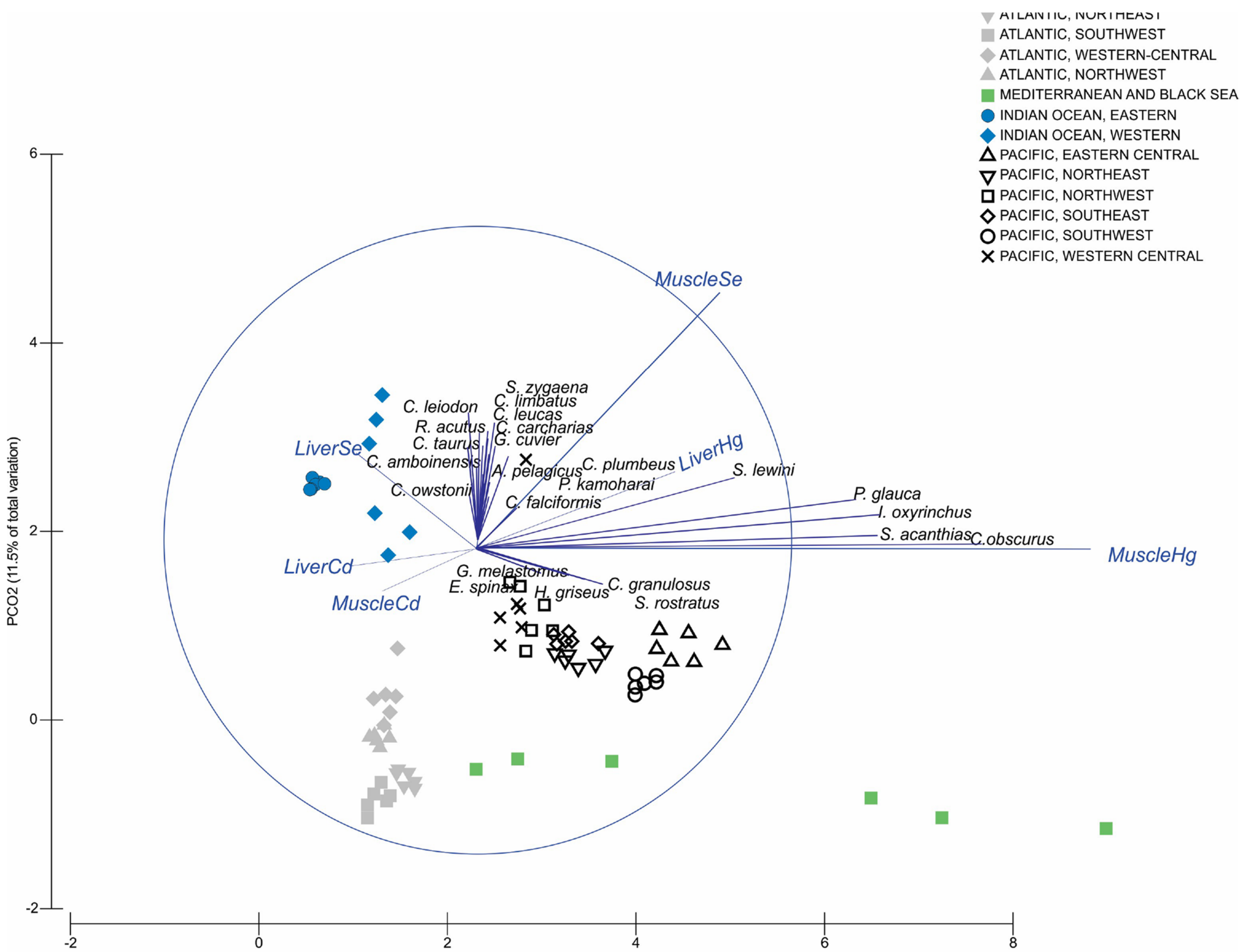

Fig. 2 Principal Coordinates Analysis (PCO) comparing mean concentrations of Hg, Se and Cd in muscle and liver of sharks from different oceanic regions. The end of the vectors denote the highest concentration of a given element/tissue

to the relation Sq.Dist/SD were always four: *I. oxyrinchus, S. acanthias, P. glauca* and *C.obscurus. S. lewini* was also quite important being one of the species responsible for these differences four out of five times, although for the difference between Hg in muscle with Se in liver, this species was not important. These results are graphically depicted in Fig. 2, the PCO. The horizontal axis better explains the concentration of Hg in both liver and muscle, as well as the concentrations of Cd in muscle and liver. The concentrations of Hg are higher in the Mediterranean and Black Sea, as well as in the Eastern Central Pacific (Mexican Pacific, Hawaii, Gulf of California). The shark species identified with SIMPER can be seen in this horizontal axis to the right; it is evident that the higher Hg concentrations in muscle of *S. acanthias, C. obscurus* and other species from the Mediterranean, Black Sea and the Eastern Pacific influenced these results. To the left, the horizontal axis explains the concentration of Cd in both tissues and Se in liver, which is higher in the western Indian ocean (East Africa) and the Atlantic for a considerable number of species.

FISH ASSEMBLAGES. A total of 358 studies were examined ranging from the year 1971 to 2018, and 45 studies not indicating the year in which they were undertaken. The majority of studies were done after the year 2000, particularly after 2010 (Fig. 3a). Elemental concentrations were reported mostly in muscle, followed by liver and fins. There was a tendency to increase the number of published papers in recent years (after 2007) for muscle and liver; in the case of fins, reports started on 2008. These studies comprised 109 shark species from a wide range of habitats (pelagic, demersal, benthopelagic, bathy demersal, reef associated). The TP of the analyzed organisms ranged from 3.4 to 4.7, from mesopredators to top predators, although most of the organisms had a TP above 4, indicating that the majority of the studied species were top predators (Fig. 3b). The frequency of published papers presenting Hg, Se and Cd in sharks was arranged for muscle (Fig. 3c) and liver (Fig. 3d) along the period of the review (fins were not included because of the low number of studies). As can be seen, muscle has been more studied than liver. With respect to the elements, Hg has been more studied than the other elements, perhaps because of the toxicological relevance of Hg and the general interest associated to the elevated dietary human intake of Hg through shark consumption.

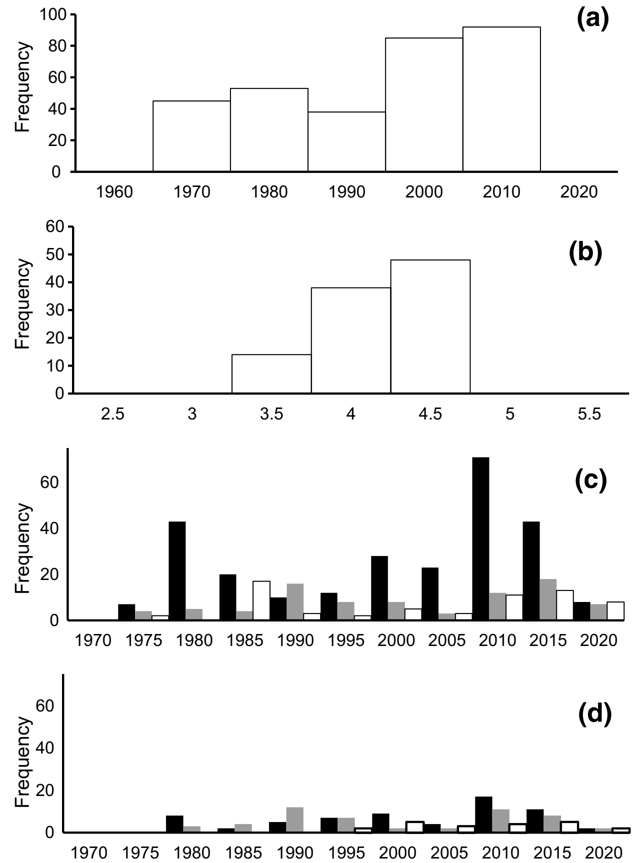

Fig. 3 Frequency of studies analyzing the decade in which the studies were made (**a**), and the trophic levels of shark species (**b**). Frequency of studies analyzing concentrations of Hg (black bars), Se (white bars) and Cd (gray bars) in muscle (**c**), and in liver (**d**) of sharks in different years (Color figure online)

Baseline Levels

The determination of natural background levels in aquatic habitats as a relative measure to distinguish between natural concentrations and anthropogenically influenced concentrations is complicated and highly challenging (Solaun et al. 2013). However, recently, the baseline metal(oid) concentrations in marine organisms have been modeled using probability frequency distribution. The criteria is that the baseline levels is operationally defined as the 5% distribution (Lu et al. 2019). The cumulative frecuency distributions of the three elements in the two tissues of sharks conformed to the logistic model (Figs. 4 and 5). The S-curve of the models exhibited different distribution patterns for different elements and the two tissues. The determination coefficients for all elements in the model ranged between 0.882 for Cd in liver, to 0.984 for Hg and Se in muscle, consistent with the behavior of the cumulative frequency distribution. It is important to highlight that there are very limited empirical studies addressing the cumulative distribution in global or entire geographic environments (Lu et al. 2019).

Previous studies (Méndez-Fernández et al. 2017; Rodriguez et al. 2018) suggested that 90th percentile values of metals in biological tissues discriminate toxic and non-toxic sites, which can be regarded as an ecological threshold tissue concentration (ETTC). Though Lu et al. (2019) indicate that using baseline estimates (based on the 90th percentile values) in tissues over enormous geographical scale can achieve to misinterpretations. Theoretically, baseline concentrations are generated from unaltered nature conditions as estimates

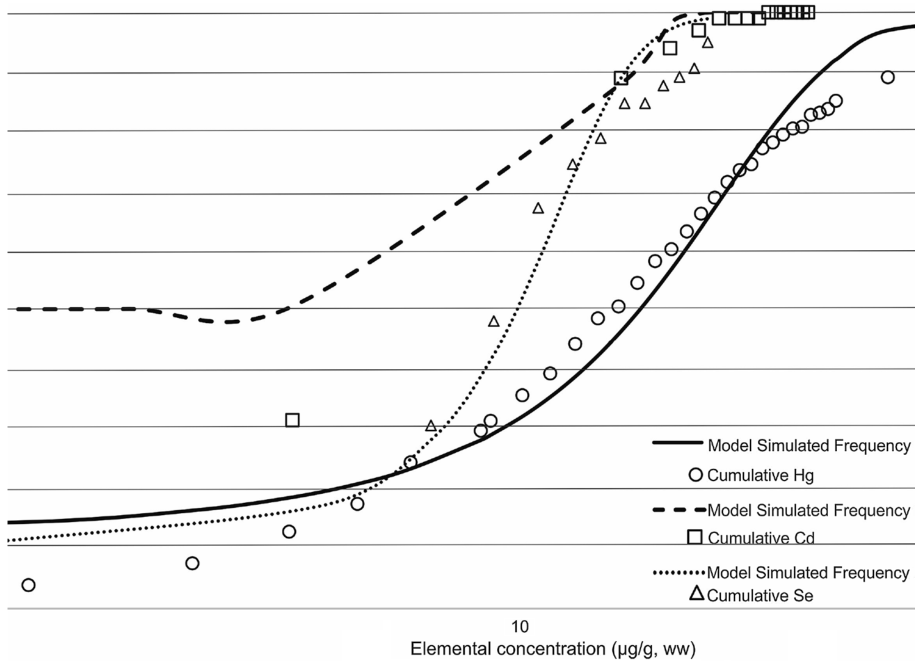

Fig. 4 Cumulative frequency distributions and baselines of Hg, Cd and Se (μg g^{-1} wet weight) in muscle of sharks from all analyzed FAO regions. Baselines for muscle were: Hg 15% = 1, 20% = 5.3, 50% = 17.4, 90% = 37.4, 95% = 50; Cd 15% = 0, 20% = 0, 50% = 0.00014, 90% = 11.44, 95% = 8.7; Se 15% = 0.7, 20% = 1.4, 50% = 2.8, 90% = 5.1, 95% = 5.9

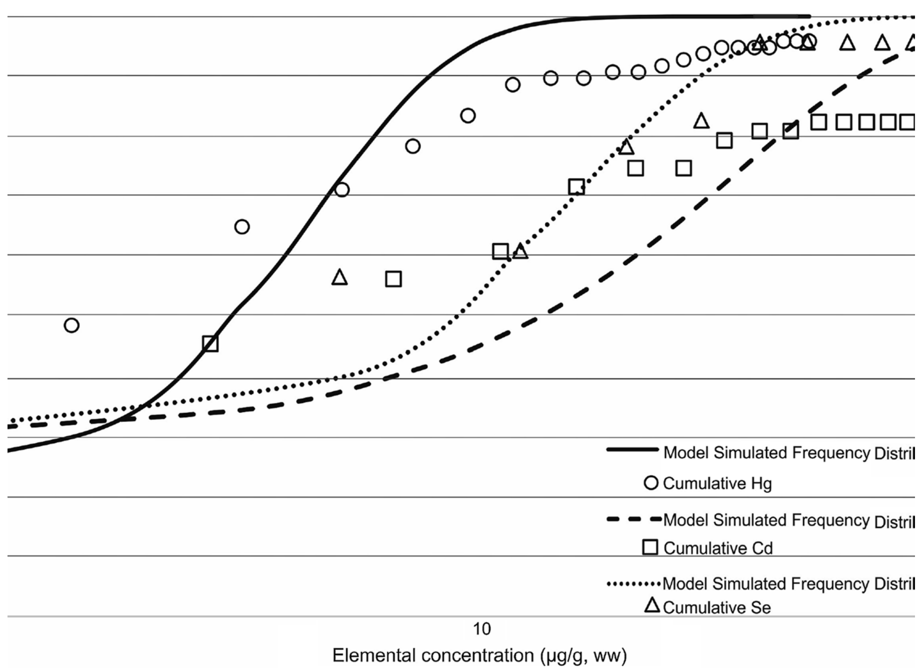

Fig. 5 Cumulative frequency distributions and baselines of Hg, Cd and Se (μg g^{-1} wet weight) in liver of sharks from all analyzed FAO regions. Baselines for liver were: Hg 15% = 0, 20% = 0, 50% = 3.8, 90% = 7.7, 95% = 9.5; Cd 15% = 0, 20% = 0, 50% = 13.6, 90% = 37.5, 95% = 51.1, Se 15% = 0, 20% = 0, 50% = 9.2, 90% = 20.6, 95% = 25.7

of the no-observed-effect concentrations (NOEC), which are much lower concentrations than ETTC. Based on the cumulative frequency analysis of metals and metalloids, Lu et al. (2019) proposed the 5th percentile values ($C_{5\%}$) as the baseline concentration. Thus, the calculated tissue baseline 5% element concentrations (wet weight basis) in muscle (Fig. 4) and liver (Fig. 5) of sharks were: for Hg, 0.129 μg/g in muscle, 0.147 μg/g in liver; for Cd, 0.517 μg/g in muscle, 0.290 μg/g in liver; for Se, 0.105 μg/g in muscle, 0.218 μg/g in liver. Baseline levels were calculated for the sharks in general, and another one for the species in which there were sufficient data to examine individually. In some cases, the baseline levels (μg g^{-1} wet weight) for mercury in muscle of sharks were directly correspondent to the levels registered in tissues of sharks from the oceanic regions with low Hg atmospheric deposition flux (< 10 μg m^{-2} y^{-1}; Fig. 1): sites from the west section of the Pacific Eastern Central (0.007, 2010; 0.05, 1990), and the Atlantic southwest (0.0106, 0.0137, 0.0185, no year of collection). However, similar baseline levels (µg g^{-1} wet weight) of 0.01 and 0.03 during the decade of 2010´s were found in the western Indian ocean (Persian Gulf), and baseline levels of 0.034 and 0.048 during 2010 were found in the Pacific Southeast at sites where the Hg atmospheric deposition flux is > 10 µg m^{-2} y^{-1}.

In the case of Hg in liver, a baseline of 0.0000001 (µg g^{-1} wet weight) was found in the Pacific Eastern Central, where the atmospheric deposition flux of Hg is low (< 10 µg m^{-2} y^{-1}; Fig. 1). However, baselines of 0.02 (µg g^{-1} wet weight) were found in areas with high Hg atmospheric deposition flux (> 10 µg m^{-2} y^{-1}), Indian Ocean Eastern and Western, the Mediterranean and Black Sea, and the Atlantic Southwest.

The baseline for Cd in muscle and liver was 0.00005 (µg g^{-1} wet weight), which appeared in 22 studies from different regions, although the majority of these studies were undertaken at the Altantic Northeast and Pacific Northwest. For Se in muscle, baseline levels (µg g^{-1} wet weight) corresponded to the Atlantic Western Central and Atlantic Northeast (0.05 and 0.07, both from the years 2000´s). For Se in liver, the baseline also corresponded to the Atlantic Northeast with values of 0.4, both from 2010.

For the specific case of the most studied species, the baseline of Hg (µg g^{-1} wet weight) in muscle for *P. glauca*, was 0.048 from the Pacific southeast, for *I. oxyrinchus* it was 0.034 also from the Pacific southeast, for *S. lewini* it was 0.55 from the Pacific eastern central, for *S. zygaena* it was 0.25 also from the Pacific eastern central, for *G. melastomus* it was 0.74 from the northeast Atlantic, and for *S. acanthias*, the baseline was 0.07 from the Atlantic Northeast.

Implications for the Human Health

Legal limits of studied elements in fish and fishery products consumed by humans vary among countries; common limits of Cd (0.5 µg g^{-1}; Nauen 1983), Hg (1.0 µg g^{-1}; CEC 2006) and Se (1.0 µg g^{-1}; Nauen 1983) were compared with average concentrations for all the species. Levels of Cd in muscle were above the legal limits in 6% of the reports; in the case of Hg, concentrations in muscle were above the legislation in 49% of the reports; for Se, in 25% of the studies the levels were above the limits considered in the legislation. As can be seen, Hg was the element that was surpassed in more sharks than Cd and Se. This issue is of concern since Hg might affect consumers.

As it has been documented, fish consumption is the main source of Hg and other elements to humans. In this context, the HI provides an indication of the non-cancer risk to consumers depending on the rate of fish consumption and the levels of the elements in the edible portion of sharks. The HI was estimated considering all studies whose species reported Cd, Hg and Se levels in muscle tissue and the corresponding HQ's; as expected, HQ and HI values were highly variable. Considering all the studies with elemental concentrations in muscle tissue, values of HQ for Cd (0.10 ± 0.18) and Se (0.11 ± 0.10) were lower than for Hg (2.05 ± 2.69). The HI was above one (2.26), this indicates that considering average concentrations of Cd, Hg and Se in muscle tissue of all sharks there might be a risk during the lifetime of an individual different to cancer. It is worth mentioning that the estimation was made considering an average weight for an adult of 62 kg and an individual rate of shark consumption of 41.9 g day^{-1}; even though not all shark species are usually consumed, HI and HQ estimations were made assuming that all reported shark species are used for human nutrition. Such values of average weight for an adult and shark consumption are very variable among the different regions of the planet and they were used just as a first approximation. Since mean values of HQ were higher than one only for Hg, results of HQ in FAO Regions are presented (Table 3); with the exception of HQ_{Hg} (0.21 ± 0.27) in FAO region Pacific Southeast, all FAO regions had HQ_{Hg} above one. It is noticeable that the highest HQ_{Hg} (5.35 ± 5.99) in the region named as Mediterranean and Black Sea coincides with one of the regions with the highest atmospheric deposition of Hg (Fig. 1).

Considering the risks associated to Hg, it has been suggested that given the antagonistic effect of Se over Hg, it is more realistic to consider the occurrence of both elements in the edible portion of fish. Such interaction may be useful to assess the benefit or risk to consumers. Mass and molar concentrations of Hg and Se and corresponding estimations of HBV_{Se} are presented in Table 4. Considering average molar concentrations of Hg and Se, Se levels are higher than Hg; such relationship resulted in an average HBV_{Se} of positive value (3.06 ± 13.84), it indicates that

Table 3 Estimated hazard quotients for Hg (HQ_{Hg}) in muscle of all shark species according to FAO region

FAO region	*n*	HQ_{Hg}
Mediterranean and Black sea	21	5.35 ± 5.99
Atlantic western central	35	2.29 ± 1.97
Pacific eastern central	40	2.22 ± 3.42
Pacific western central	35	2.13 ± 3.06
Indian Ocean western	41	2.01 ± 1.45
Indian Ocean eastern	24	1.82 ± 1.57
Atlantic northwest	7	1.44 ± 1.15
Pacific southwest	34	1.40 ± 0.93
Pacific northwest	14	1.38 ± 1.37
Atlantic southwest	25	1.30 ± 1.26
Pacific northeast	4	1.12 ± 0.32
Atlantic northeast	17	1.10 ± 0.75
Pacific southeast	3	0.21 ± 0.27
Global		2.05 ± 2.69

n number of shark species with reported Hg concentrations in muscle shark consumption improves Se levels in humans and is beneficial. As can be observed, the elevated variability of HBV_{Se} values means that there are some cases with negative results; *i.e.,* consumption of those sharks implies a risk for consumers. Considering FAO Fishing areas, negative HBV_{Se} values corresponded to Mediterranean and Black Sea (− 48.37), Atlantic Northwest (− 5.54), Pacific Eastern Central (− 0.97), and Atlantic Southwest (− 0.21). As can be seen, two areas are of concern (Mediterranean and Black Sea, Atlantic Northwest) since HBV_{Se} indicate potential health risks associated to shark consumption. On the contrary, in seven FAO regions the HBV_{Se} values were positive (range 0.25–16.15) so no health risk for consumers exist. Information related to measurements of Hg and Se in the edible portion of sharks is necessary, especially in regions (Indian Ocean Eastern, Pacific Northeast, Pacific Southeast) where HBV_{Se} has not been estimated.

Conclusion

A detailed analysis and relationship between shark landings and studies could not be done accurately, because, other than the blue shark, the world statistics data on shark landing is very poor, with the majority of countries not reporting information by species, but only by groups, i.e., shark landings and ray landings, including all the species. However, it seems that the number of studies on Cd, Hg and Se in sharks is related to the commercial importance of certain species, as is the case of the blue shark, which is the most landed species in the world, according to FAO (2015), and it is also the species with more studies.

There is also a strong relationship between the number of studies and certain regions and countries which have a strong scientific community. Most of the studies come from developed countries and with well-known fisheries research programs, such as New Zealand, Australia, the USA, Canada, Europe and Japan. There are few studies from Latin America and the Middle East, and none (at least published) from Africa. Therefore, the studies published in relation to this subject reflect the interest for commercially important species, especially from developed countries.

Differences between metal concentrations and trophic position were not found, because the reported TP for the majority of the species is above 4. These TP values were obtained from fishbase (Froese and Pauly 2021), and it is known that they are usually inaccurate (Amezcua et al. 2015). The problem is that studies analyzing concentrations of trace metals seldom do analyses of the trophic positions of the analyzed organisms and do not report their lengths. The consequence is that the TP that can be obtained in fishbase, if it exists, is usually for adult organisms. It is known that many shark species have different feeding habits through their lives, which implies that the TP is different too. However, a comprehensive study including feeding behavior, ontogenetic changes in diet and TP, and concentration of trace metals in sharks would be needed to obtain information of the relationship between trace metal concentration and TP.

There was no difference in the concentrations of metals among habitats (demersal, benthic, pelagic, oceanic, etc.), and time (years). The reason for this is likely to be related to the fact that in most of the cases the studies do not document accurately either where the sharks were caught, so the habitat considered is that found in the references, however, it is known that species such as hammerhead sharks, which is

Table 4 Mass (µg g^{-1} wet weight) and molar (µmol kg^{-1}) concentrations of Hg and Se, and corresponding Se health benefit value (HBV_{Se}) in muscle of sharks from the different FAO regions

		Mass		Molar		
FAO region	*n*	Hg	Se	Hg	Se	HBV_{Se}
Mediterranean and Black sea	1	12.15	3.24	60.57	41.03	− 48.37
Atlantic western central	3	1.48 ± 1.62	1.24 ± 1.08	7.36 ± 8.07	15.66 ± 13.71	9.11 ± 15.99
Pacific eastern central	11	1.65 ± 1.51	0.86 ± 1.01	8.24 ± 7.52	10.87 ± 12.85	− 0.97 ± 18.61
Pacific western central	17	0.93 ± 0.55	0.77 ± 0.35	4.62 ± 2.72	9.76 ± 4.37	6.67 ± 5.56
Indian Ocean western	4	1.54 ± 1.92	1.57 ± 1.17	7.67 ± 9.60	19.90 ± 14.77	16.15 ± 10.14
Indian Ocean eastern	0	–	–	–	–	–
Atlantic northwest	1	1.11	0.27	5.53	3.42	− 5.54
Pacific southwest	8	1.13 ± 0.35	0.92 ± 0.78	5.61 ± 1.73	11.61 ± 9.86	6.28 ± 13.63
Pacific northwest	6	0.20 ± 0.07	0.42 ± 0.14	1.01 ± 0.34	5.32 ± 1.79	5.29 ± 1.95
Atlantic southwest	2	0.84 ± 1.05	0.55 ± 0.29	4.21 ± 5.25	7.00 ± 3.72	− 0.21 ± 13.87
Pacific northeast	0	–	–	–	–	–
Atlantic northeast	13	0.85 ± 0.61	0.52 ± 0.29	4.23 ± 3.04	6.57 ± 3.71	0.25 ± 11.73
Pacific southeast	0	–	–	–	–	–
All regions	65	1.23 ± 1.66	0.81 ± 0.74	6.13 ± 8.29	10.20 ± 9.33	3.06 ± 13.84

n number of shark species with reported Hg and Se concentrations in muscle considered circumglobal and pelagic, have different habitats through their lifespan, even as adults. In relation to the time, the concentrations have remained similar, but it is also necessary to consider that there are many gaps in the studies. This could influence the results obtained.

Most of the elemental concentrations in FAO areas were different among them, however similar concentrations were found in the south hemisphere, from western Australia (Eastern Indian Ocean), to the southwest Pacific (Pacific southamerica).

Although it may be questionable to establish baseline concentrations for sharks, individually or globally, because hypothetically these values should be obtained from pristine areas, we propose for the first time global baseline values for muscle and liver of sharks. In favor of this, is the fact that most of the proposed levels coincide (in order of magnitude) with the concentrations quantified in oceanic regions with a low atmospheric deposition flux of Hg ($< 10\ \mu g\ m^{-2}\ y^{-1}$).

Implications for human health by the consumption of shark were evaluated considering the legal limits, the hazard index (HI) and the hazard quotient (HQ), and the Se health benefit value (HBV_{Se}). When elemental concentrations in muscle were contrasted with the legal limits, it was found that Hg is of concern since its values exceeded the legal levels in 49% of the studies. HQ and HI values were highly variable, in muscle HQ for Cd (0.10 ± 0.18) and Se (0.11 ± 0.10) were lower than for Hg (2.05 ± 2.69). HI was 2.26, which indicates that all sharks there might be a risk during the lifetime of an individual different to cancer. The elevated variability of HBV_{Se} values (negative) indicates that consumption of certain sharks implies a risk for consumers. Considering FAO Fishing areas, two areas are of concern (Mediterranean and Black Sea, Atlantic Northwest). On the contrary, in seven FAO regions the HBVSe values were positive so no health risk for consumers exist. In this context, it is clear that more information related to Hg and Se in sharks is required, particularly in the Indian Ocean Eastern, Pacific Northeast, and Pacific Southeast, where HBVSe could not be calculated.

Supplementary Information The online version contains supplementary material available at https://doi.org/10.1007/s44169-021-00006-2.

Acknowledgements Thanks are due to Carlos Suárez for figure preparation.

Funding Funding was obtained from the Ministry of Public Education of Mexico (Project TecNM 7596.20-P).

Declarations

Conflict of interest The authors declare that they have no conflict of interest.

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