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Baseline

Arsenic contamination in widely consumed Caribbean sharpnose sharks in southeastern Brazil: Baseline data and concerns regarding fisheries resources

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ABSTRACT

Although shark meat is consumed worldwide, elevated arsenic (As) concentrations have been increasingly reported. The Caribbean sharpnose shark (*Rhizoprionodon porosus*) is a widely consumed fishing resource in Brazil, with scarce information on As burdens to date. Herein, commercial-sized juvenile Caribbean sharpnose sharks from Rio de Janeiro (Brazil) were assessed in this regard, presenting significantly higher hepatic As concentrations in males (8.24 ± 1.20 mg kg^{-1} wet weight; $n = 12$) compared to females (6.59 ± 1.87 mg kg^{-1} w.w.; $n = 8$), and a positive correlation ($r = 0.74$) was noted between female muscle and liver As concentrations, indicating systemic inter-tissue transport not evidenced in males. Arsenic concentrations were over the maximum permissible As limit established in Brazilian legislation for seafood and calculated Target Hazard Quotients suggest consumption risks, although cancer risks were not evidenced. Therefore, Public Health concerns with regard to Caribbean sharpnose shark As burdens deserve careful attention.

Food is both a biological necessity and a human right supported by article 25 of the Universal Declaration of Human Rights since 1948, and, in Brazil, by the Federal Constitution (Conti and Schroeder, 2013). However, according to the World Health Organization (WHO), the ingestion of food items contaminated by bacteria, viruses, parasites or chemical substances is responsible for the death or illness of about one in ten people each year worldwide, characterizing significant Public Health risks and increasing annual public health costs (Kite-Powell et al., 2008; Vandermeersch et al., 2015; WHO, 2015).

Metals and metalloids have been increasingly detected at high concentrations in the environment, due to their historical use in diverse human activities (Martorell et al., 2011; Vandermeersch et al., 2015). These are among the most important anthropogenic chemical pollutants, due to their environmental persistence and the ability to bioaccumulate and, in some cases, biomagnify along trophic webs (Kite-Powell et al., 2008; Olmedo et al., 2013), in several aquatic organisms, such as fish (Omar et al., 2013; Whyte et al., 2009). Exposure to metals and metalloids can, thus, also reach and result in harmful effects in humans. However, each element acts differently in different organisms, and toxicity depends on numerous factors, such as exposure duration, dosage and chemical form (Järup, 2003).

Arsenic (As) is a low abundance metalloid in the earth's crust (0.0001%), found in the marine environment in both its inorganic form, mainly in anoxic environments, and in its organic form, as a result of

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aquatic organism decomposition or excretion (Akter et al., 2005; Oremland and Stolz, 2003). Due to its high toxicity risks, As is included in the World Health Organization's list of the ten most toxic substances to human health (WHO, 2021).

The main human exposure route to As is through water and food ingestion. The As present in drinking water is a significant source of exposure to the inorganic and most toxic form of this element and has led to a high number of contamination cases in several extremely populous South and Southeast Asia regions (Singh et al., 2015; WHO, 2001). Following water contamination, food exposure, including seafood, is generally the main contributor to total As daily intakes (WHO, 2001). However, although inorganic As is the predominant As species in both freshwater and marine environments, organoarsenic species, generally considered non-toxic, are the main forms detected in seafood (Francesconi, 2010; Saavedra et al., 2008), generally comprising 85 to 98% of total As content in these food items (Borak and Hosgood, 2007; Feldmann and Krupp, 2011). This is due to the fact that inorganic As is rapidly biotransformed in fish into methylarsenic and other organic As species (Bergés-Tiznado et al., 2021).

Inorganic As, although the most toxic form of this element is also the least representative in fish, as stated previously, usually ranging from 1 to 4% (Bergés-Tiznado et al., 2021; Borak and Hosgood, 2007), although some authors have reported values up to 10% (De Gieter et al., 2002; FSANZ, 2019; Marengo et al., 2018; Souza-Araujo et al., 2021). While less studied, some organic As species are also considered to be conceivably harmful, such as fat-soluble As, arsenosugars and other organoarsenicals (e.g., fat-soluble and lipophilic arsenic compounds), which, due to lack of information on their toxicity, should be reported as potentially toxic arsenicals (Feldmann and Krupp, 2011; Taleshi et al., 2010).

Bioaccumulation and biomagnification processes are expected to occur in marine ecosystems, although data concerning As is controversial in the literature. It is known that As concentrations can be controlled by excretion. Therefore, As bioaccumulation, in general, reflects the exposure duration to this element, thus reflecting a balance between incorporation and excretion. Some literature assessments have reported data concerning As biomagnification and biodilution processes, which can occur between trophic levels, as well as during species growth (Bergés-Tiznado et al., 2021; Vieira et al., 2020). Nevertheless, it is interesting to note that a meta-analysis concerning five trophic level marine food webs from 33 countries and regions demonstrated that As assimilation and enrichment is species-specific and a global trend towards biodilution (Sun et al., 2020). This suggests that inorganic As contents usually decrease, whereas arsenobetaine (AsB) burdens increase throughout trophic levels, as species belonging to higher trophic levels display greater ability to biotransform inorganic As into the less toxic AsB (Bergés-Tiznado et al., 2021; Sun et al., 2020).

Due to food safety and, consequently, Public Health concerns, several studies have been carried out regarding As, raising concerns regarding high total As concentrations in seafood worldwide (Borak and Hosgood, 2007; WHO, 2019). In this regard, marine organisms can contain total As burdens ranging from less than 1 to over 100 mg kg^{-1}, mainly as organic As species, i.e., arsenosugars in macroalgae and AsB in invertebrates and fish (WHO, 2001). This is, however, still poorly monitored and the commercialization of As-contaminated fish remains a worldwide concern (WHO, 2011), including in developed countries, such as the USA, where about 90% of the total As consumed originates from seafood (Borak and Hosgood, 2007).

Different countries and regions have adopted different ways of regulating As levels in seafood, to avoid possible human health risks. In Brazil, for example, the Brazilian National Health Surveillance Agency (ANVISA), responsible for food contaminant control, defines the legal maximum limit for total As as 1 mg kg^{-1} wet weight (w.w.), in fish (ANVISA, 2013), while Venezuela has established a limit of 0.1 mg kg^{-1} w.w. and Hong Kong, of 10 mg kg^{-1} w.w. (Torres et al., 2017). The WHO (2019), however, has recently banned a previously established maximum permissible limit of 1.0 mg kg^{-1} and now considers that no level of As intake is completely safe for human consumption, as exposure to this metalloid may lead to significant Public Health implications, depending on the amount of contaminated seafood consumed (Horta et al., 2011; Kite-Powell et al., 2008).

In Brazil, fishing was one of the first productive activities to be established in the country (Abdallah and Sumaila, 2007), representing an important source of income in several regions (Clauzet et al., 2005). In this regard, artisanal (traditional) fisheries represent over 50% of the national fishery production, reaching up to 70% in some areas (Begossi et al., 2011). Fishing is, therefore, an important economic activity in many Brazilian municipalities, with the income obtained through these activities comprising a significant part of local budgets (Diegues, 2008). However, some studies have reported that total As concentrations in the marine biota of several Brazilian coast areas are relatively high (Angeli et al., 2013; Catharino et al., 2008), exceeding the maximum established limits still in place for seafood in the country (ANVISA, 2013).

Shark meat is an important source of protein in several regions worldwide (Simpfendorfer et al., 2011). In Brazil, this fish category, sold under the generic name "*cação*", is highly appreciated, mainly due to affordable prices and a lack of bones and the fact that this is one of the two fish categories legally purchased for school meals in public schools in the state of Rio de Janeiro (Rio de Janeiro, 2005; SEEDUC, 2019). However, it is important to note that several shark species are actually long-lived top predators vulnerable to contaminant bioaccumulation and biomagnification processes, which may, ultimately, become harmful to human health through their consumption (Adel et al., 2016; Gilbert et al., 2015).

In this context, the Caribbean sharpnose shark (*Rhizoprionodon porosus*) comprises an important fishing resource for the entire Brazilian fish production chain and is widely consumed throughout the country, especially in the state of Rio de Janeiro, notably in Cabo Frio region. These sharks are small coastal marine sharks distributed in tropical regions, usually found near the coast, continental, and island shelves of the Caribbean and South America, although they can be found in offshore waters up to 500 m in depth. They are considered top predators, feeding mainly on small bony fish, but also some invertebrates, such as squid and shrimp (Compagno, 1984). Currently, Caribbean sharpnose shark populations are categorized as vulnerable, with a decreasing trend and possible population reductions ranging from 30 to 49%, in the last 14 years, due to the current levels of widespread exploitation of these organisms, by both artisanal and industrial fisheries, which generally lack adequate management, particularly in the southwest Atlantic (Carlson et al., 2021). In addition to fisheries overexploitation, coastal contamination by metals and metalloids, especially in more urbanized regions, can threaten elasmobranch health and pose Public Health risks (Martins et al., 2021). Because of this, studies concerning As concentrations in this group, are paramount, especially in southeastern Brazil. Hence, this study aimed to provide baseline data regarding total As burdens in these highly consumed sharks in southeastern Brazil.

The *Rhizoprionodon* members comprise the main genus captured in fisheries in the highly productive Cabo Frio region, in the state of Rio de Janeiro, southeastern Brazil (FIPERJ, 2019). Twenty juvenile Caribbean sharpnose shark, eight females and 12 males, were purchased at the Pontal de Santo Antônio fishmarket, in Tamoios (−22.597147 S, −41.994951 W), located in the district of Cabo Frio, Rio de Janeiro state, Brazil (Fig. 1). The Caribbean sharpnose sharks were purchased from an artisanal fisherman's stall (Fig. 2).

The coast of the state of Rio de Janeiro, and especially the Cabo Frio region, displays a strong fishing tradition, due to a local upwelling phenomenon that enriches continental shelf waters in nutrients, providing an important food source for several marine organisms and, consequently, high abundance of fishery resources (Coelho-Souza et al., 2012; Valentin, 2001). This upwelling mainly affects the region comprising the coast of the municipality of Cabo Frio, which, with a population of 226,525 people (IBGE, 2017), displays significant fishing

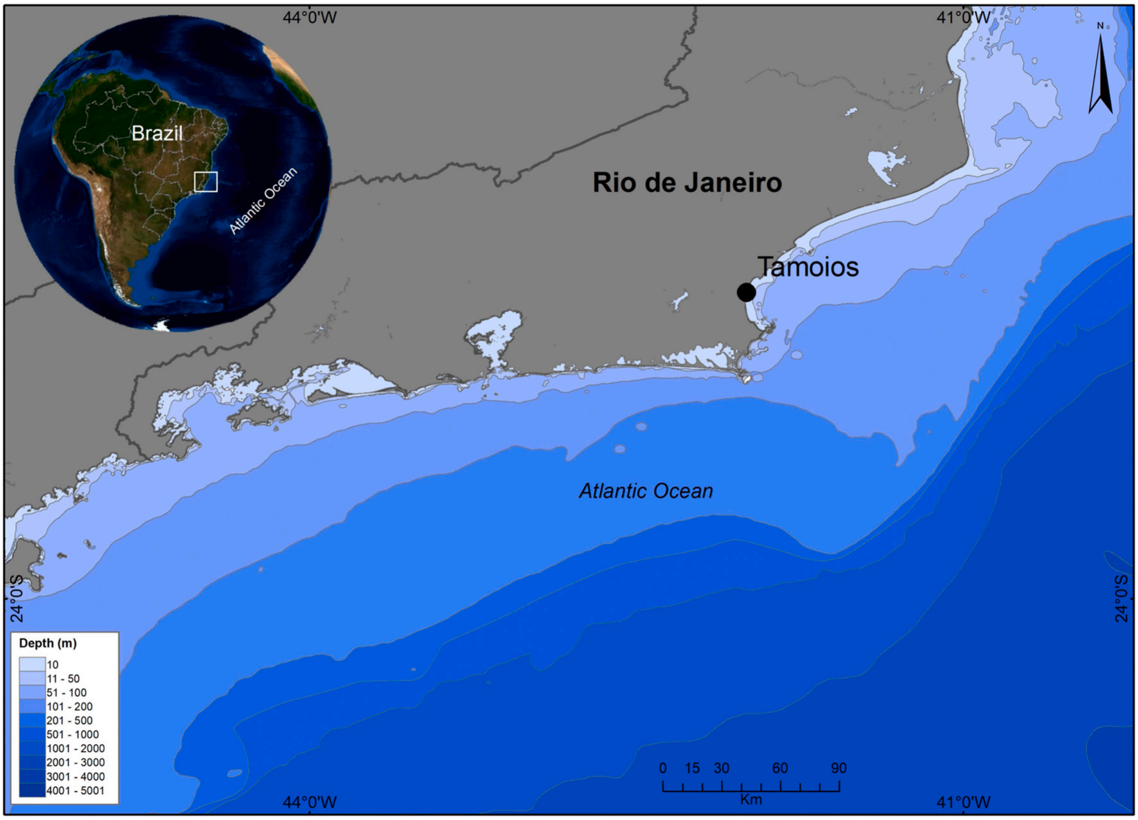

Fig. 1. Map of the Caribbean sharpnose shark (*Rhizoprionodon porosus*) sampling site, in the district of Tamoios, Cabo Frio, Rio de Janeiro state, southeastern Brazil.

Fig. 2. Caribbean sharpnose sharks (*Rhizoprionodon porosus*) for sale at Cabo Frio, southeastern Rio de Janeiro. Source: Hauser-Davis Research Group collection.

activity, both artisanal and industrial. This intense fishing activity makes Cabo Frio one of the four main fishing ports in the state of Rio de Janeiro, with a monthly average of 701.39 t of fish landed in 2018 and in the first half of 2019 (FIPERJ, 2018a, 2018b, 2019).

The Caribbean sharpnose sharks were transported on ice in Styrofoam boxes to the laboratory. Total weights for each specimen were obtained using a balance (0.01 g) and total length (distance from the tip of the snout to the largest end of the caudal fin) were determined to the nearest centimeter using a measuring tape. The specimens were then dissected, and liver and muscle aliquots were transferred into sterile 15 mL polypropylene tubes and frozen at −20 °C until the analyses.

After thawing, about 100 mg of each sample were weighed, transferred to new sterile 15 mL polypropylene tubes, mixed with 1.0 mL of bidistilled nitric acid (HNO_3, 67% v/v) and left to stand overnight. The following day, the samples were heated to 100 °C in the capped tubes for 4 h, avoiding As losses (USP, 2013). After reaching room temperature, the samples were adequately diluted with ultra-pure water (resistivity >18.0 MΩ cm) obtained from a Merck Millipore water purifying system (Darmstadt, Germany), for subsequent analysis by inductively coupled plasma mass spectrometry (ICP-MS) employing a NexIon 300× spectrometer (PerkinElmer, USA).

Method accuracy was verified by the parallel analysis of procedural blanks and of a certified reference material (ERMBB422 - fish tissue, European Commission). Certified reference material recovery values were considered adequate for this type of study (Observed value: 13.4 ± 4.6 mg kg^{-1}; Certified value: 12.7 ± 0.7 mg kg^{-1}, recovery: 106%), as per Eurachem standards (Eurachem, 1998; Ishak et al., 2015). Multi-elemental external calibration was applied by appropriate dilutions of a mixed standard solution (Merck IV) and non-spectral interferences were corrected using ^{103}Rh as the internal standard introduced online through a capillary prior to the beginning of the analysis, fixed at 20 mg L^{-1}, and remaining throughout the entire analysis. All determinations were performed in triplicate. Analytical curve correlation coefficients were always above 0.995. The limit of quantification (LOQ) was calculated according to the Brazilian National Institute of Metrology, Quality and Technology (INMETRO, 2016), as LOQ = (10*SD*df)/slope of the line, where SD is the standard deviation of the ratio of the analytical signal to the internal standard signal of 10 blanks and df is the applied dilution factor, as 0.088 mg kg^{-1}.

Considering the daily intake of a specific element, it is possible to estimate the maximum safe amount of food items that can be consumed without risk of harmful effects, using a reference dose termed the Provisional Tolerable Weekly Intake (PTDI). This parameter comprises the maximum level of exposure to a certain contaminant for a certain period of time, as established by the joint FAO/WHO committee (WHO, 2011). Herein, this was calculated for muscle tissue intake of the investigated Caribbean sharpnose shark specimens, and the obtained values were compared to the reference value proposed by the WHO for this element.

To accurately investigate potential Public Health concerns, the most representative value regarding fish intake in the study area should be considered. In this regard, Brazil displays an average per capita fish intake of 24.74 g a day, or 173.18 g per week (MPA, 2012). Therefore, the average As value detected in the shark muscle tissues were multiplied by the average Brazilian fish consumption and divided by the average body weight of a Brazilian adult, estimated at 70 kg (MPA, 2012). The highest detected As value in shark muscle was also used in the same calculation for a worse-case scenario assessment.

The target hazard quotient (THQ), developed by the United States Environmental Protection Agency (US EPA, 1989), is often applied to verify potential risks associated to fish consumption (Ramos-Miras et al., 2019). It is defined as the exposure ratio to a certain toxic element and an established reference dose comprising the highest level at which no adverse health effects are expected. This approach was applied to assess non-carcinogenic health risks from Caribbean sharpnose shark consumption. When THQ <1, non-carcinogenic health effects are not expected, and when THQ >1, risks concerning adverse health effects are probable. The THQ was calculated as:

$$THQ = (E_{FR} \times E_D \times F_{IR} \times C) / (RfD \times BWa \times ATn) \times 10^{-3}$$

where E_{FR} denotes the exposure frequency (days per year, totaling 365), E_D comprises the exposure duration (equivalent to an average lifetime of 76.3 years for the Brazilian population (IBGE, 2019)), F_{IR} consists in the food ingestion rate in grams per day for the respective food item, C is defined as the concentration of the analyzed element in the investigated food item, as wet weight, R*f*D is the oral reference dose of the element, in μg g^{-1} day^{-1} (set at 0.0003 for As), BWa denotes average body weight (estimated at 70 kg per adult), ATn is the averaged exposure time to the toxic element (365 days*76.3 years) and 10^{-3} is the unit conversion factor.

The target cancer risk (TCR), used to assess potential risks associated with exposure to carcinogenic agents throughout a lifetime exposure period, was also calculated, as As is a carcinogenic element, using an oral slope factor that determines the probability of excess cancer risk over the lifetime of the exposed individual (Antoine et al., 2017), as:

$$TCR = (E_{FR} \times E_D \times F_{IR} \times C \times CPS_O) / (BWa \times ATc) \times 10^{-3}$$

where E_{FR} denotes the exposure frequency (days per year, 365), E_D comprises the exposure duration (equivalent to average lifetime of 76.3 years for the Brazilian population (IBGE, 2019)), F_{IR} is the food ingestion rate in grams per day for the respective food item, C is the concentration of the carcinogenic element in the given food item, in wet weight, CPS_O is the oral cancer slope factor for inorganic arsenic, set at 1.5 mg kg^{-1} day^{-1}, BWa denotes average body weight (estimated at 70 kg per adult), ATc is the averaged exposure time to the carcinogen (365 days*76.3 years) and 10^{-3} is the unit conversion factor.

Data normality was verified by the Shapiro-Wilk W test. As the data were normally distributed, parametric tests were subsequently applied. The Student *t*-test was used to verify potential differences between male and female Caribbean sharpnose shark muscle As concentrations, as well as between liver and muscle. Potential correlations between As concentrations in Caribbean sharpnose shark muscle and liver were assessed by Pearson's correlation test, categorized according to Bryman and Cramer (2012) as very weak when $0.00 < r < 0.19$; weak when $0.20 < r < 0.39$; moderate when $0.40 < r < 0.69$; strong when $0.70 < r < 0.89$; and very strong when $0.90 < r < 1.00$. A significance level of $p < 0.05$ was accepted, and all analyses were performed using the GraphPad Prism version 8.4.2 for macOS.

Table 1 presents the Caribbean sharpnose shark total As concentrations (range, means, standard deviation (SD)) and 10% inorganic As estimates as reported in other seafood assessments (FSANZ, 2019; Marengo et al., 2018), including sharks (Souza-Araujo et al., 2021), following US EPA (2000) recommendations for human health risk assessments.

All Caribbean sharpnose sharks analyzed herein were juveniles, with females ranging from 43 to 52 cm in total length, averaging 48 cm and males from 44 to 56 cm, averaging 50 cm. These sizes are commonly sold for human consumption in the study area. No significant differences ($p > 0.05$) were observed between males and females regarding both size and total As burdens in muscle. On the other hand, a significant difference between total hepatic As concentrations were observed between sexes, with higher concentrations detected in males. No significant differences comparing liver and muscle concentrations for either sex was noted. Although the difference between tissues was non-significant, five sharks (three females and two males), totaling 25% of studied samples, presented higher As burdens in muscle compared to liver.

In general, metals and metalloids are usually present in higher concentrations in the liver (Suñer et al., 1999), as this organ is primarily responsible for organism detoxification processes (Storelli and Marcotrigiano, 2004) and considered a target organ for these contaminants (Barrera-García et al., 2013). Therefore, under optimum conditions, muscle tissue tends to accumulate low metal and metalloid concentrations due to lower metabolic activities compared to the liver (Squadrone et al., 2013; Uluturhan and Kucuksezgin, 2007). Concerning As, following exposure through the dietary and environmental routes, this element enters the bloodstream and is partially metabolized by the liver, while what remains and is not eliminated via detoxification processes such as the metallothionein detoxification route, may bioaccumulate in muscle tissue (Adel et al., 2018; Land et al., 2018). In this regard, our findings concerning higher As concentrations in 25% of the muscle samples when compared to liver suggest potential As bioaccumulation. However, future studies should be carried out to confirm bioaccumulation and biomagnification processes in Caribbean sharpnose sharks from this Brazilian region.

Intra-body metal and metalloid interactions depend on several physicochemical and biotic factors, including environmental chemical speciation. This may lead to differential elemental tissue distribution patterns in animal tissues (Peterson et al., 2009; Yang et al., 2010). Associations between the same element in different organs indicate inter-tissue transport (Erasmus, 2004; Hauser-Davis et al., 2020) and are important to assess element toxicodynamics. Herein, the Pearson correlation test indicated a strong positive correlation ($r = 0.74$) between As concentrations in female Caribbean sharpnose shark muscle and liver, indicating systemic inter-tissue transport (Erasmus, 2004; Hauser-Davis et al., 2020). No correlations between muscle and liver in males or between total length and As concentrations in muscle and liver were observed. Contrasting literature information concerning this type of correlation is noted. In general, studies report no relationship between As concentrations in liver and fish size. However, most studies of this kind have been performed on bony fish. In elasmobranchs, As tends to be regulated by a more efficient excretion processes, as the metabolism of this element in the liver of these animals occurs through the methylation of inorganic As, resulting in quicker excretion of organic As forms (Storelli and Marcotrigiano, 2004). However, additional studies are paramount to further investigate this process in cartilaginous fishes.

Two other studies have been carried out concerning As burdens in *Rhizoprionodon* sharks, namely *R. porosus* and *R. terraenovae*, from Brazil and Mexico, where highly variable and relatively high As concentrations were observed compared to other assessments (Table 2).

Comparing *R. porosus* data, Souza-Araujo et al. (2021) reported a total As range between 2.19 and 15.39 mg kg^{-1} w.w., with a mean concentration of 9.58 mg kg^{-1} w.w. in the muscle of five animals, not discriminating between females and males, from the Brazilian Amazon Coast, in northern Brazil. These results were slightly higher than the

Table 1
Total arsenic and of the 10% estimated for inorganic As, in mg kg^{-1}, in wet weight, in the muscle and liver of Caribbean sharpnose sharks (*Rhizoprionodon porosus*) sampled from Cabo Frio, southeastern Brazil.

Tissue	As (range)		As (means ± SD)		10% (range)		10% (mean ± SD)	
	Females	Males	Females	Males	Females	Males	Females	Males
Muscle	4.00–10.4	3.96–12.8	6.32 ± 2.41	7.21 ± 2.10	0.40–1.04	0.40–1.28	0.63 ± 0.24	0.72 ± 0.21
Liver	3.30–8.85	5.67–10.1	6.59 ± 1.87	8.24 ± 1.20	0.33–0.89	0.57–1.01	0.66 ± 0.19	0.82 ± 0.12

SD = standard deviation.

Table 2
Arsenic (As) concentrations, in mg kg^{-1} wet weight (ww), in *Rhizoprionodon* sharks species. Total length (TL) range and means are also displayed in centimeters (cm).

Reference	Organism	Location	*n*	TL (range)	TL (mean)	Tissue	As (range)		As (mean ± SD)	
							Female	Male	Female	Male
	***Rhizoprionodon* genus**									
This study	*R. porosus*	Brazil	20	43–56	49	Muscle	4.00–10.4	3.96–12.8	6.32	7.21
						Liver	8.30–8.85	5.67–10.1	6.59	8.24
Souza-Araujo et al. (2021)	*R. porosus*	Brazil	5	np	np	Muscle	2.19–15.39		9.58 ± 5.27*	
Núñez-Nogueira (2005)	*R. terraenovae*	Mexico	21	9.6–90.9	np	Muscle	n.p.		3.68*	
						Liver	n.p.		3.27*	
Souza-Araujo et al. (2021)	*R. terraenovae*	Brazil	5	np	np	Muscle	1.61–9.30		5.96 ± 3.13*	

TL = Total length; n.p. = not provided; * = no distinction between males and females; SD = standard deviation.

mean reported herein, of 6.86 mg kg^{-1} w.w., ranging from 3.96–12.8 mg kg^{-1} w.w., when grouping males and females, 6.23 mg kg^{-1} w.w. when reporting females only and 7.21 mg kg^{-1} w.w. for males only. All values for both assessments in Brazil were above the maximum permissible limit of 1 mg kg^{-1} according to Brazilian legislation.

The mean total As concentration in the muscle tissue of the *Rhizoprionodon teranovae*, a species belonging to the same genus as the sharks investigated herein, also contained similar burdens, of 3.68 mg kg^{-1} w.w. in muscle and 3.27 mg kg^{-1} w.w. in liver in individuals sampled from the Gulf of Mexico (Núñez-Nogueira, 2005), and 5.96 mg kg^{-1} w.w. in the muscle from specimens sampled from the Brazilian Amazon Coast (Souza-Araujo et al., 2021).

Concerning Public Health risks, all individuals presented total As muscle burdens above the limit established by Brazilian legislation of 1 mg kg^{-1} w.w. (ANVISA, 2013). When taking into account the conservative value of 10% estimated as the toxic inorganic As content, the maximum permissible limit should be, in fact, 0.1 mg kg^{-1} w.w., and all samples in this study would still be above this limit, indicating potential human health risks (Marengo et al., 2018; US-EPA, 2000). Arsenic toxicity, however, is directly related to its oxidation state following environmental interactions, as well as to the amount absorbed by the body (Akter et al., 2005; Cullen and Reimer, 1989), which also affects its bioavailability and distribution (Bernhard et al., 1986). For example, changes in pH, temperature, salinity, sulfide and iron ion concentrations may result in the occurrence of As (III) and As (V), highly toxic inorganic As species (Julshamn et al., 2012; Wakao et al., 1988). In addition, as mentioned previously, inorganic species can also be biotransformed in higher trophic level marine species (Bergés-Tiznado et al., 2021; Sun et al., 2020). Therefore, speciation assessments are also important and should be implemented in this type of assessment, to indicate differences between As species, especially since the aforementioned limit is very conservative, which could put most of the Brazilian fisheries at risk, causing severe social impacts and generate even more conflict for vulnerable populations that directly or indirectly depend on fishing (Diegues, 2008; Marengo et al., 2018).

With regard to potential Public Health concerns, some agencies (ANVISA (2013), WHO (2011) and the FSANZ (2019)), suggest that when the inorganic As concentrations are unknown, exposure calculation factors can be used. Thus, dietary As intakes of Caribbean sharpnose sharks from Cabo Frio analyzed herein were calculated for comparisons to the provisional tolerable weekly intake (PTWI) for total As, inorganic As, and organic As concentrations established (0.1 mg day^{-1} inorganic arsenic per kg body weight) by the Food and Agriculture Organization of the United Nations (FAO) and the Joint FAO/WHO Expert Committee on Food Additives (JECFA) (WHO, 2011), which have now been recanted. Table 3 indicates the estimated weekly levels of arsenic exposure for the mean and worse-case scenarios for Caribbean sharpnose shark intake.

In both the mean and worse-case scenarios, inorganic As contents were above the recently removed limit by the WHO and the Brazilian ANVISA limit, indicating significant health risks to the human population that consumes these animals. It is interesting, however, that the Australia and New Zealand Food Standards (FSANZ, 2019), despite assuming a 10% inorganic arsenic to total As ratio, considers that assessments concerning dietary exposures to inorganic As should be based on actual inorganic As data, and not on the use of total As conversion factors, as inorganic As burdens can vary widely in foodstuffs, usually ranging from 1 to 4%, but exceeding 10% in some cases, due to several factors including life stage and feeding habits, among others (Bergés-Tiznado et al., 2021; Borak and Hosgood, 2007; De Gieter et al., 2002; Marengo et al., 2018; Souza-Araujo et al., 2021). Therefore, inorganic As levels may, in fact, be overestimated for several foodstuffs.

It is important to note that the Joint FAO/WHO Expert Committee on Food Additives (JECFA) establishes a provisional tolerable weekly intake (PTWI) of 0.015 mg kg^{-1} bw $week^{-1}$ as the limit for inorganic As (Uneyama et al., 2007; WHO, 2011). Furthermore, several European countries, as well as the USA, Canada and the Republic of Korea, report a daily intake of total As ranging from 0.17–3.33 μg kg^{-1}, assuming a body weight of 60 kg, much lower than the results for the Caribbean sharpnose sharks analyzed herein (WHO, 2011). However, according to

Table 3
Mean and worse -case scenarios for the Caribbean sharpnose sharks concerning arsenic contents in muscle tissue in specimens from Cabo Frio.

As species	Estimated level of exposure (mg kg^{-1} bw $week^{-1}$)
Mean As scenario	
Total As	16.96
Organic As	15.26
Inorganic As	1.69
Worse-case As scenario	
Total As	31.75
Organic As	28.57
Inorganic As	3.17

Uneyama et al. (2007), in relation to organic As, the ingestion of about 0.05 mg kg^{-1} bw day^{-1} or less does not seem to trigger adverse effects.

The target hazard quotient (THQ) was calculated as 8.08 for the mean value scenario and 15.08 for the worse-case scenario concerning Caribbean sharpnose shark consumption, both using total As concentrations. This indicates severe non-carcinogenic health risks concerning the consumption of these sharks for As. However, as discussed previously, the most toxic As fraction is inorganic. Therefore, the same calculations were performed using 10% inorganic As, resulting in 0.81 and 1.50 for the mean value and worse -case scenarios, near and over 1, respectively, still indicating concerns. The target cancer risk (TCR) was calculated as 3.94×10^{-9} for the mean value scenario and 7.38×10^{-9} for the worse-case scenario concerning Caribbean sharpnose shark consumption concerning total As concentrations, and as 5.05×10^{-8} and 9.44×10^{-8} concerning inorganic As concentrations, respectively. Since the range for acceptable risk of developing cancer is reported as 10^{-6} to 10^{-4} (Shaheen et al., 2016; US-EPA, 1991), consumption of the evaluated sharks does not exceed the 10^{-4} cancer threshold and is not a cause for concern for cancer development. However, the mean consumption for fish items in Brazil does not take into account vulnerable populations such as children, the elderly and pregnant women, who are vulnerable due to aging, low body weight or due to the pregnancy process (US-EPA, 2021; Wiech et al., 2021). Furthermore, it does not consider traditional communities, such as fishers and quilombolas (traditional communities descended from escaped slaves), who may ingest higher seafood amounts than the national means. Therefore, calculated TCRs for each specific group should be implemented to assess realistic cancer risks. However, it is important to note that Brazil lacks epidemiological studies on the occurrence of food transmitted diseases, including chemical contamination by As in fish (Carmo et al., 2005; Secretaria de Vigilância em Saúde, 2021; Silva, 2013). Consequently, epidemiological monitoring requires significant improvements, especially concerning notifying the occurrence of As contamination due to fish consumption, producing robust, centralized and updated data.

This study indicates Public Health concerns regarding Caribbean sharpnose shark consumption in southeastern Brazil, due to high total As concentrations over the maximum permissible limit established in the country, even more so when taking into account the recent removal of the maximum limit for this element by the WHO, as it was not considered health-protective for exposed human populations. Furthermore, the target hazard quotient (THQ) even applying the conservative 10% inorganic As burden was of near and over 1 for the mean value and worse-case scenarios, respectively, also indicating concerns. The TCR was lower than the range for acceptable risk of developing cancer, although this calculation does not consider specific vulnerable populations such as the elderly, children, and pregnant women, as well as fishers and quilombolas. Even so, potential overestimations of risks are still noted, as As contents in sharks may not reach the 10% inorganic amount, as they have been reported as mostly ranging from 0.6% to 5% total As (Souza-Araujo et al., 2021). There is, therefore, an urgent need for speciation assessments in this regard, which are even more necessary considering that fisheries are a significant economic activity in Brazil and other developing countries, and care must be taken to ensure both seafood safety and the livelihood of many vulnerable populations.

CRediT authorship contribution statement

Oswaldo Luiz de C. Maciel: Data Curation, Visualization, Investigation, Formal analysis, Writing - Original Draft, Writing – Draft reviewing; **Isabel Q. Wilmer**: Data Curation, Visualization, Investigation, Formal analysis, Writing - Original Draft; **Tatiana D. Saint'Pierre**: Validation, Resources, Funding acquisition, Supervision, Writing - Original Draft; **Wilson Machado**: Conceptualization, Resources, Investigation, Validation, Data Curation, Formal analysis; Project administration, Supervision, Writing - Original Draft, Writing – Draft reviewing.; **Salvatore Siciliano**: Conceptualization, Resources, Funding acquisition, Project administration, Supervision, Writing - Original Draft, Writing – Draft reviewing; **Rachel Ann Hauser-Davis**: Conceptualization, Resources, Investigation, Validation, Data Curation, Formal analysis; Project administration, Supervision, Writing - Original Draft, Writing – Draft reviewing.

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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