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Mercury Status of the Amazon Continental Shelf: Guiana Dolphins (*Sotalia guianensis*, Van Benédén 1864) as a Bioindicator

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Abstract Total mercury (Hg) was analyzed in muscle tissue of 27 accidentally captured Guiana dolphins (*Sotalia guianensis*) in order to evaluate Hg contamination levels present in the Amazon Continental Shelf, in Amapá state, North Brazil. The samples showed a mean concentration of 0.4 ± 0.16 µg/g wet weight (ww), ranging from 0.07 to 0.79 µg/g ww. As observed in several other cetacean species, Hg concentrations presented positive correlations to body length, related to the capacity to bioaccumulate this element throughout life. Hg concentrations were not significantly different between males (mean = 0.38 µg/g ww; n = 15) and females (mean = 0.42 µg/g ww; n = 12). Concentrations were low when compared to results of studies carried out with small cetaceans in the Northern Hemisphere, and with some previous studies in the southeastern region of Brazil. In contrast with high Hg concentrations normally detected in river dolphin samples from Amazon River tributaries, our results suggest that the Amazon coast contains low levels of Hg in bioavailable form.

Keywords Guiana dolphins · *Sotalia guianensis* · Amazon shelf · Mercury · Brazil

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In the last several decades, a considerable increase in the number of chemical industries has been observed. This has resulted in the presence of increased amounts of many chemical pollutants, including mercury (Hg), in the marine environment, mainly in coastal regions. This trend has impacted environmental integrity, biodiversity and human health (Fleming et al. 2006; Moura et al. 2011a). There is considerable evidence that background levels of mercury in the environment have increased in recent time. This evidence comes from studies of lakes and ocean sediment profiles and ice cores, and from various types of samples collected in remote areas, as well as the industrialized regions of the planet (Outridge et al. 2007). Most wildlife are exposed to Hg primarily as MeHg through the diet, rather than to other chemical forms of Hg. Due to its lengthy persistence and high mobility in the ocean, MeHg shows a high level of biomagnification in the upper levels of the food web (O'Shea 1999).

Mercury may cause multiple symptomatic effects, such as neurochemical, reproductive, behavioral, physiological, immunological and histological changes, affecting the health and survival of the mammals that are exposed, including coastal dolphins that are chronically exposed (Scheuhammer et al. 2007). High concentrations of Hg have been detected in sediment, fishes, river dolphins and humans from the Amazon Basin (River) (Rosas and Lehti 1996; Lailson-Brito et al. 2008; Passos and Mergler 2008; Hacon et al. 2009).

The presence of Hg in the Amazon Basin (River) has been attributed to anthropogenic activities (e.g. gold mining; deforestation process; hydroelectric plants) or natural sources (e.g. Hg-rich soil), and is known as a historical problem for the humans that rely on fish as their main sources of protein (Passos and Mergler 2008; Hacon et al. 2009). On the other hand, in the Amazon Continental Shelf, low levels of Hg have been detected in sediment, fish, crabs and human samples (Patchineelam 2001; Vilhena et al. 2003); and it is important to understand how this element biomagnifies in apex marine predators, such as Guiana dolphins (*Sotalia guianensis*) that are very common in the region (Siciliano et al. 2008).

Cetaceans have been considered a bioindicator species for the health of the marine environment, mainly due to their biological characteristics and feeding habits (Aguilar et al. 1999). These marine mammals are top predators in the marine trophic web, and have long life-spans. Therefore, they are highly exposed to persistent contaminants, like MeHg, through bioaccumulation and biomagnification processes (O' Shea 1999). Due to its nearshore distribution (<50 m deep), the Guiana dolphin (*S. guianensis*) is especially vulnerable to the effects of the pollution in coastal waters along its distribution from Santa Catarina, Brazil, to Nicaragua on the Caribbean coast (Di Beneditto and Ramos 2004) (Fig. 1). Therefore, some concerns regarding possible adverse effects of toxic chemicals on this coastal dolphin have been noted, mainly related to the emerging infectious diseases that have been detected in some populations along the Brazilian coast (Van Bressem et al. 2007; 2009). The aim of this study was to evaluate the concentrations of total mercury in muscle tissues of Guiana dolphins accidentally captured on the Amazon Continental shelf, located in the south region of Amapá state, northern Brazil.

Materials and Methods

The sampling area is located along the southern coast of Amapá state, northern Brazil, into the Amazon Continental Shelf (Fig. 1). The Amazon Shelf, which is located between the Pará estuary and approximately 5°N and between the coast and 100 m isobath, has a broad inner shelf (about 250 km in width), an elevated middle shelf (40–60 m depth), and an outer shelf extending to the shelf break at the 100 m isobath (Nittrouer and DeMaster 1996).

The circulation on the Amazon Shelf caused by the complex interaction of strong tidal currents, river discharge, wind force, and the flux of the North Brazil Current, that flows close to the shore (Nittrouer et al. 1991). The Amazon shelf is characterized by a mixture of river and ocean waters, great abundance of energy and complex biological and physiological processes mainly associated

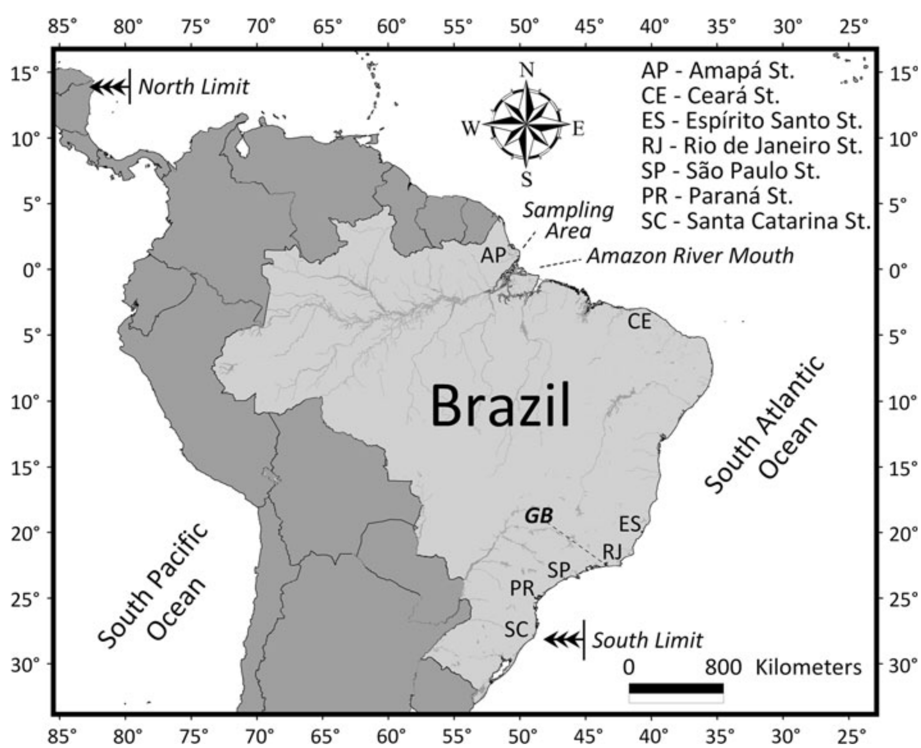

Fig. 1 Sampling area located on the Amazon Continental Shelf, southern Amapá state, northern Brazil. *Arrows* showing the northern (Nicaragua, Caribbean Sea) and southern (Santa Catarina, Southern Brazil) limits of the distribution of Guiana dolphins (*S. guianensis*). The abbreviations refer to the states mentioned in the text and other sites cited, such as Guanabara Bay (GB)

with extensive fluvial and sedimentary discharge from the Amazon River. The water discharge of the Amazon River into the continental shelf ranges from 100,000 to 220,000 m^3 s^{-1}, and the solid discharge from 11 to 13 × 10^8 tons $year^{-1}$ (Kineke et al. 1996).

Muscle samples were collected from 27 Guiana dolphins accidentally captured in fishing nets during 2007 by the Amapá fishing fleet during a fish monitoring program. Fifteen dolphins were males and 12 were females. The total body length varied from 126 to 192 cm (mean = 169) and 148 to 192 cm (mean = 175) for males and females, respectively (Table 1). The dolphins caught were measured and sexed inside the fishing boats. Muscle samples were collected from the dorsal region of the fresh dead dolphins immediately behind the dorsal fin. The samples were collected with sterilized material and were immediately stored at −20°C. Necropsies of each dolphin were performed for the collection of biological material, biometric measurements and sex determination according to the Geraci and Lounsbury (2005) protocol. The body length of the animals was measured along the longitudinal axis of the body, from the tip of the upper jaw to the caudal notch. All samples were collected in fresh carcasses, a few hours after death (code 2; Geraci and Lounsbury 2005).

Approximately 0.5 g of tissue was weighed, and digested with a 1:1 mixture of sulfuric and nitric acid, in the presence of 0.1 % vanadium pentoxide (Vega et al. 2009). The digestion temperature was 80°C, and oxidation was completed by the addition of a sufficient volume of 5 % m/v potassium permanganate solution. Immediately before instrumental analysis, the oxidant excess was reduced with a 20 % m/v solution of hydroxylammonium chloride, diluted to 50 mL. Mercury levels were determined using cold vapor atomic absorption spectrometry (Techtron, Model AA-5, Varian Instrument Division, Palo Alto, CA) with a detection limit being 0.05 μg/L. Calibration curves were prepared using dilutions of 1,000 μg mL^{-1} standard solutions prepared from Titrisol (Merck, Darmstadt, Germany) ampoules diluted with 0.2 % v/v nitric acid. Analyte addition tests demonstrated the adequacy of this calibration procedure, as well as the lack of any multiplicative matrix effect.

Quality control was performed by a strict blank control, the analysis of replicates and certified reference materials. Accuracy was assessed through the analysis of certified material DORM-1 (Hg: 798 ± 74 ng g^{-1}) from NIST (National Institute of Standards and Technology; http://www.nist.gov). Average recovery values were always ≥90 % of the certified values. Reproducibility was evaluated using the coefficient of variation of the replicates, which was always less than 25 %.

An independent *T* test was used to test the differences between the mean metal concentrations of males and females. A simple linear regression analysis (R^2; significance at $p < 0.05$) was used to further explore the relationship between the metal concentrations as a function of total length (TL) of individuals. Statistical analyses were performed using the software Windows SPSS (http://www-01.ibm.com/software/analytics/spss/).

Results and Discussion

The presence of Hg was detected in all samples. Concentrations are expressed on a ww basis . Dry weight (dw) basis concentrations in the literature were converted to wet weight (ww) basis concentrations, assuming a moisture content of 70 % based on the results of Yang and Miyazaki (2003). The mean concentration of total Hg was 0.4 ± 0.16 μg g^{-1} ww, ranging from 0.07 to 0.79 μg g^{-1} ww (Table 1). Hg concentrations in muscle tissue remained below those detected in other studies with *S. guianensis* along the Brazilian coast (Kehrig et al. 2004; Lopes et al. 2008; Carvalho et al. 2008) (Table 2). In Guanabara Bay, Rio de Janeiro state, known as a highly polluted environment, Kehrig et al. (2004) detected an average concentration of 0.7 μg g^{-1} ww in stranded Guiana dolphins. A similar mean concentration (0.73 μg g^{-1} ww) was found in Guiana dolphins accidentally captured along the northern coast of Rio de Janeiro state (Carvalho et al. 2008). These authors also found relatively high concentrations of Hg in cutlassfish (*Trichiurus lepturus*) muscle tissue, increasing with weight and body length. This is the preferential prey

Table 1 Descriptive statistics for body length and mercury concentrations in muscle of males (M, n = 15) and females (F, n = 12) of Guiana dolphins from Amapá state, northern Brazil

	Hg concentration (μg g^{-1} ww)			Body length (cm)		
Sex	M	F	All	M	F	All
Mean	0.38	0.42	0.4	169.3	175.2	171.8
SD	0.14	0.17	0.16	18.9	13.3	16.9
Median	0.39	0.45	0.42	177	178.5	1.78
Maximum	0.66	0.79	0.79	192	192	192
Minimum	0.11	0.07	0.07	126	148	126

Table 2 Means, standard deviations and ranges of total mercury concentrations (μg g^{-1} wet weight) in muscle tissues of several small cetaceans from areas around the world

Species	Hg	Localities	n	Ref
T. truncatus	2.85 (0.63–5.1)	Portugal, Atlantic Ocean	2	a*
T. truncatus	8.9 ± 12 (0.37–39)	Israel, Mediterranean	17	b
T. truncatus	12.78 (1.59–25.5)	France, Atlantic Ocean	5	c*
T. truncatus	(0.22–0.77)	Australia	–	d
T. truncatus	100	Corsica, Mediterranean	1	e*
T. truncatus	9.55 ± 6.01 (2.36–22.5)	Japan	9	f
T. truncatus	2	South Africa	3	g*
T. gephyreus	5.5 ± 0.8	Argentina	2	h
S. longirostris	1.39 ± 0.30 (0.84–1.76)	Taiwan	9	i
S. longirostris	0.47 (0.4–0.57)	Gulf of California, Mexico	9	J*
S. longirostris	1.1 (0.87–1.33)	Sta Lucia, Caribbean	2	k
S. coeruleoalba	15.9	Italy, Mediterranean	39–51	l*
S. coeruleoalba	15 ± 27.1 (1.04–63.4)	Japan	5	f
P. phocoena	3.5 ± 14.4 (0.2–108)	Germany–Baltic and North Sea	57	m
S. attenuata	3.64 ± 2.21 (1.05–12)	Taiwan	53	i
G. macrorhynchus	9 (8.19–9.84)	New Caledonia	2	n*
S. guianensis	0.7 (0.2–2.5)	Guanabara Bay, RJ, Brazil	15	o
S. guianensis	1.8 ± 0.46	Espírito Santo state, Brazil	5	p
S. guianensis	0.73 (0.34–1.42)	Northern Rio de Janeiro state	6	q
S. guianensis	0.98	Northern Rio de Janeiro state	21	r*
S. guianensis	1.07 ± 0.35 (0.2–1.66)	Rio de Janeiro state, Brazil	20	s
S. guianensis	0.4 ± 0.16 (0.07–0.79)	Amazon coast, Brazil	27	t

Ref references

* Converted from dry weights assuming 70 % moisture content (Yang and Miyazaki 2003)

a: Carvalho et al. (2002); b: Roditi-Elasar et al. (2003); c: Holsbeek et al. (1998); d: Kemper et al. (1994); e: Frodello et al. (2000); f: Endo et al. (2003); g: Henry and Best (1999); h: Marcovecchio et al. (1990); i: Chen et al. (2002); j: Ruelas et al. (2000); k: Gaskin et al. (1974); l: Monaci et al. (1998); m: Siebert et al. (1999); n: Bustamante et al. (2003); o: Kehrig et al. (2004); p: Lopes et al. (2008); q: Carvalho et al. (2008); r: Kehrig et al. (2009); s: Moura et al. (2011b) t: this study

species of *S. guianensis,* and seems to be a potential contributor to the bio-transference of Hg for this coastal dolphin. Pantoja (2001) analyzed the stomach contents of 23 Guiana dolphins collected in the same region where the dolphins of the present study were collected. According to this author, Guiana dolphin males prefer fish, principally *T. lepturus* while females feed on fish and shrimp. Juvenile dolphins feed predominantly on shrimp.

Mercury concentrations in dolphin muscle in the present study indicate that the levels are low compared to concentrations observed in small cetaceans from other regions in the world (Table 2). As examples, Roditi-Elasar et al. (2003) detected a high mean concentration (8.9 ± 12 μg g^{-1} ww; n = 17) of Hg in bottlenose dolphins (*Tursiops truncatus*) from the Israeli coast of the Mediterranean Sea. Marcovecchio et al. (1990) detected a mean concentration of 5.5 ± 0.8 μg g^{-1} ww (n = 2) in bottlenose dolphins (*Tursiops gephyreus*) from the coast of Argentina. In addition, Endo et al. (2003) also detected high levels of Hg for muscle tissue in striped dolphins (*Stenella coerulealba*) sampled along the coast of Japan, with a mean of 15 ± 27.1 (n = 05). Hepatic Hg concentrations in small cetaceans, including *S. guianensis*, have indicated values similar to those in cetaceans from highly polluted areas in the northern hemisphere, such as the Mediterranean Sea and Asia (Kunito et al. 2004).

Few studies have been conducted on the concentrations of metals in marine organisms from the northern coast of Brazil (Andrade and Patchineelam 1999; Lima et al. 2006). Siqueira (2003) analyzed the concentrations of zinc, cobalt and nickel in sediment samples from the Amazon continental Shelf, and attributed the low concentrations to the natural presence of these elements in the studied region. Similar results are present in Santos and Faria (1993), who evaluated the concentrations of metals (Fe, Mn, Co, Cr, Cu, Ni, Zn, Pb) in sediments in the coastal region of Amapá state. The authors suggested that the metals analysed are associated with the natural mineral composition (feldspar, mica and plagioclase) in this region that act as a geochemistry support for these elements. According to Neto and Berrêdo (2008) there are no anthropogenic influences along the Amazon coast of Brazil that could justify the presence of Hg in high concentrations in biological samples of marine organisms and sediment. Vilhena et al. (2003) found low levels of total Hg in samples of sediment, crabs (*Ucides cordatus*) and human hair collected in a mangrove swamp and surroundings located in the northeast coast of Pará state, on the Amazon coast. Contrasting with our results, Lailson-Brito et al. (2006) detected an extremely high hepatic concentration of Hg in one tucuxi dolphin (*Sotalia fluviatilis*; 215.9 µg g^{-1} ww) and in four Amazon River dolphins (*Inia geoffrensis,* 35.9 µg g^{-1} ww) collected in large tributaries of the Amazon Basin (Negro, Japurá and Madeira rivers). According to Lailson-Brito et al. (2008), the high Hg levels detected in *S. fluviatilis* are probably associated with its bioavailability from the Negro river and an adjacent water body. Gold mining activities have been conducted in the three mentioned rivers, although in minor scale in the Negro river (Lailson-Brito et al. 2008). Our results compared to the levels of Hg detected in aquatic animals from the Amazon basin suggests that this metal is not carried in large quantities or in a bioavailable form to the coast in the Amazon River flux. The previous studies of metals in sediment, fish, crabs and human samples in the Amazon coast have corroborated this hypothesis (Santos and Faria 1993; Siqueira 2003; Vilhena et al. 2003; Andrade and Patchineelam 1999; Lima et al. 2006). However, the dilution effect of metals caused by the large amount of transported water and particulate matter, as well as the differences in physico-chemical parameters between the river and coastal areas, may play an important role in the observed differences in Hg.

It is widely accepted that Hg levels are positively related to the size and age of dolphins. Confirming this trend, a smooth positive and statistically significant ($p < 0.05$) correlation between Hg concentration and body length was also observed in males ($R^2 = 0.318$) and females ($R^2 = 0.427$) of *S. guianensis* specimens from this study (Fig. 2). A similar pattern of Hg accumulation with body length was observed in Guiana dolphins from the coast of Ceará state (Monteiro-Neto et al. 2003) and from the northern coast of the Rio de Janeiro state (Carvalho et al. 2008). Also, a positive relationship between tissue Hg concentration and age in *S. guianensis* was observed in dolphins collected from São Paulo and Paraná states (Kunito et al. 2004). The relationship between metal accumulation and cetacean size may be a consequence of a higher capability to bioacumulate elements throughout their life span than to eliminate them. The higher positive relationship of Hg and length in females suggest that the transference and elimination of body concentration to calves through milk and placenta is not significant in *S. guianensis*, as already observed in other species of dolphins. According to Itano et al. (1984) the proportion of the gestational transference of Hg levels to the fetus is only 0.4–1 % of the maternal burden in striped dolphins (*Stenella coeruleoalba*). These authors also found very low levels being transferred through milk to the newborns.

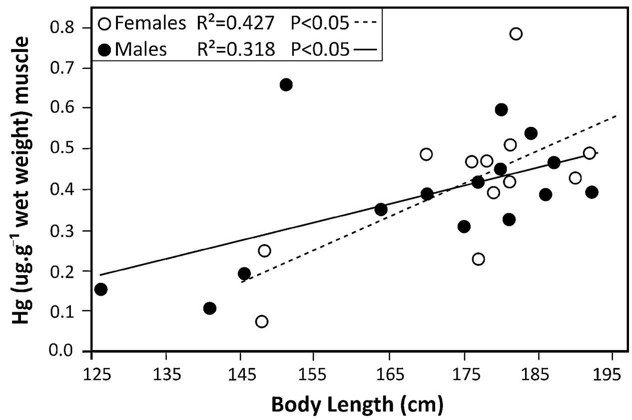

Fig. 2 Relationship between body length and Hg concentrations in muscle tissue of male and female Guiana dolphins (*S. guianensis*) from the Amazon Continental Shelf, Southern Amapá state, northern Brazil

The positive relationship between Hg and size in muscle of males is due the bioaccumulative capability of this tissue throughout the lives of the dolphins. In addition, as the size of the prey and the quantities of food ingested tend to increase in proportion to the growth of the dolphins, trophic supplies of the metal may also progressively increase (Monteiro-Neto et al. 2003).

No statistical differences ($p > 0.05$) were observed in Hg levels regarding gender. Mean Hg concentrations were quite similar, being 0.41 ± 0.17 and 0.40 ± 0.14 µg g^{-1} for females and males respectively. In general, no significant differences in Hg concentrations have been observed in tissues of females and males of marine mammals (O'Shea 1999). This same lack of variation in Hg concentrations by gender in Guiana dolphins has been observed in other studies (Monteiro-Neto et al. 2003; Kunito et al. 2004; Seixas et al. 2009; Moura et al. 2011b).

The present work adds new information on Hg concentrations in muscle tissue of a small coastal dolphin from the Amazon coast.

Low concentrations of total Hg were found in muscle tissue of Guiana dolphins when compared with other small cetaceans worldwide, and also with studies carried out with the same species in Brazil. As observed in several other cetacean species, Hg concentrations presented positive relationships to body length, likely related to the capacity to bioaccumulate this element throughout life. Mercury concentrations were not significantly different between males and females. In contrast with high Hg concentrations normally detected in river dolphin samples from Amazon

River tributaries, our results indicate that the Amazon coast contains low levels of Hg in a bioavailable form.

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