Chemosphere 262 (2021) 127819

Contents lists available at ScienceDirect

Chemosphere

journal homepage: www.elsevier.com/locate/chemosphere

Influence of sex, size and trophic level on blood Hg concentrations in Black caiman, *Melanosuchus niger* (Spix, 1825) in French Guiana

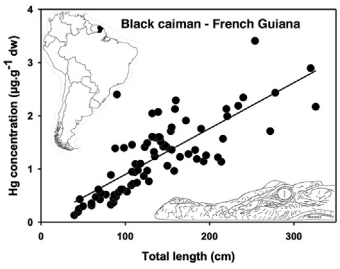
Jérémy Lemaire [a, b, *], Paco Bustamante [b, c], Olivier Marquis [d], Stéphane Caut [e, f], François Brischoux [a]

[a] Centre D'Etudes Biologiques de Chizé (CEBC), UMR 7372 CNRS-La Rochelle Université, 79360, Villiers en Bois, France
[b] Littoral Environnement et Sociétés (LIENSs), UMR 7266 CNRS-La Rochelle Université, 2 Rue Olympe de Gouges, 17000, La Rochelle, France
[c] Institut Universitaire de France (IUF), 1 Rue Descartes, 75005, Paris, France
[d] Parc Zoologique de Paris, Muséum National D'Histoire Naturelle, 53 Avenue de Saint Maurice, 75012, Paris, France
[e] Consejo Superior de Investigaciones Cientificas (CSIC), Departamento de Etologia y Conservacion de La Biodiversidad - Estacion Biologica de Doñana - C/ Americo Vespucio, S/n (Isla de La Cartuja), E-41092, Sevilla, Spain
[f] ANIMAVEG Conservation, 58 Avenue Du Président Salvador Allende, F-94800, Villejuif, France

HIGHLIGHTS

- Total mercury concentration is determined in Black Caimans in French Guiana.
- Mercury concentration increases with individual body size.
- Mercury concentration increases with their trophic position $\delta^{15}N$

GRAPHICAL ABSTRACT



ARTICLE INFO

Article history:
Received 9 April 2020
Received in revised form
20 July 2020
Accepted 23 July 2020
Available online 3 August 2020

Handling Editor: Chennai Guest Editor

Keywords:
Crocodilians
Trace metals
Tropical ecosystems
Diet
Bioaccumulation

ABSTRACT

Mercury (Hg) is a contaminant that is impacting ecosystems worldwide. Its toxicity is threatening wildlife and human populations, leading to the necessity of identifying the most affected ecosystems. Therefore, it is essential to identify pertinent bioindicator organisms to monitor Hg contamination. In this study, we determined the stable carbon ($\delta^{13}C$) and nitrogen ($\delta^{15}N$) isotope ratios in the red blood cells (RBCs), and the total Hg concentration in total blood of 72 *Melanosuchus niger* in French Guiana. The goals of our study were to assess the level of Hg contamination in total blood of Black caimans and to further investigate the influence of individual traits (i.e., sex, size/age, diet) on Hg concentrations. Mercury concentration in total blood of Black caimans ranged from 0.572 to 3.408 µg g^{-1} dw (mean ± SD is 1.284 ± 0.672 µg g^{-1} dw) and was positively correlated to individual body size and trophic position ($\delta^{15}N$). We did not find any sexual or seasonal effects on Hg concentrations in the blood. The use of blood of *M. niger* is relevant to determine Hg concentrations within the population and suggests that this species can be used as a bioindicator for environmental contamination. In addition, our results emphasize trophic position as a major source of Hg variation and further suggest that it is essential to take trophic position ($\delta^{15}N$) into account for future studies.

* Corresponding author. Centre d'Etudes Biologiques de Chizé (CEBC), UMR 7372 CNRS-La Rochelle Université, 79360, Villiers en Bois, France.
E-mail address: jeremy.lemaire@univ-lr.fr (J. Lemaire).

https://doi.org/10.1016/j.chemosphere.2020.127819
0045-6535/

1. Introduction

Mercury (Hg) is one of the major contaminants that affect human and wildlife around the world with increasing levels due to anthropogenic activities (Ericksen et al., 2003; Scheuhammer and Sandheinrich, 2007; Hsu-Kim et al., 2018). In anoxic conditions, microorganisms can transform inorganic Hg into methylmercury (MeHg), the most toxic and bioavailable form of Hg (Compeau and Barta, 1985; Benoit et al., 2003). Because of its strong bioavailability, MeHg is highly absorbed and retained in biota. Hence, it accumulates within organisms during their lifetime and biomagnifies through food webs, resulting in an increasing Hg contamination level throughout most living organisms (Mason et al., 1995; Atwell et al., 1998; Power et al., 2002).

Due to their life history traits, crocodilians (caimans, true crocodiles, alligators, gharials) are potentially good bioindicators of environmental contamination. Indeed, they are long-lived predators, resulting in the accumulation of Hg over a lifespan of several decades. As ectothermic vertebrates, crocodilians display relatively low metabolic rates, but relatively high tissue conversion rates; two features that are expected to favor the bioaccumulation of significant levels of Hg (Cook et al., 1991; Camus et al., 1998; Jagoe et al., 1998; Twining et al., 1999; Schneider et al., 2015; Lázaro et al., 2015; Nilsen et al., 2017). Maternal transfer, an elimination pathway of Hg, which is already known from a variety of reptiles, can also be found in crocodilians (Day et al., 2005; Nilsen et al., 2020). However, some studies reported similar Hg concentrations in males and females, whereas females that have already reproduced should theoretically have lower concentrations than males (Burger et al., 2000; Eggins et al., 2015; Nilsen et al., 2020).

The geographic range of caimans is altered by intense gold mining, an anthropogenic activity that is a major source of Hg deposition to the aquatic ecosystems of equatorial South America, and represents approximately 70% of local Hg emissions that are increasing the availability throughout food webs (De Lacerda, 2003; Rocha et al., 2018; Ottenbros et al., 2019). Contrarily to highly mobile individuals such as migratory birds or fishes (Fréry et al., 2001; Fort et al., 2014), crocodilians are rather sedentary (Hutton, 1989; Magnusson et al., 1991; Fujisaki et al., 2014; Caut et al., 2019). Hg concentrations in their tissues reflect the contamination of their environment at a relatively small and precise spatial scale when measured in the blood. In equatorial South America, this taxon could provide the opportunity to assess environmental Hg contamination due to its wide distribution. In addition, sample collection from crocodilians is rather uncomplicated as there is sufficient tissue (i.e., blood, scutes, claws) that can be sampled with comparatively little impact on individuals.

Stable carbon and nitrogen isotope analyses provide information on the diet composition and the trophic position of organisms via the variation of $\delta^{13}C$ and $\delta^{15}N$ levels. Carbon stable isotopes ($\delta^{13}C$) are used as a proxy to discriminate different types of habitat and to provide information on primary production (Pinnegar and Polunin, 2000; Post, 2002). Nitrogen stable isotopes ($\delta^{15}N$) are discriminating the trophic position, as consumers are predictably enriched in ^{15}N in relation to their diet (Minagawa and Wada, 1984; Post, 2002; Vanderklift and Ponsard, 2003). Among crocodilians, it has been shown that increased values of $\delta^{15}N$ reflect a change in the trophic position, linked to a change in their diet (Radloff et al., 2012; Bontemps et al., 2016; Caut et al., 2019). Therefore, adding analyses of stable carbon and nitrogen isotopes to the quantification of Hg levels is extending information on the feeding habitat and trophic positions.

In this study, we assessed Hg concentrations in the only known population of Black caimans (*Melanosuchus niger*) in French Guiana (De Thoisy et al., 2006). This French territory suffers from illegal, artisanal, small-scale gold mining. The goals of our study were to determine the levels of Hg contamination in the blood of Black caimans and the factors influencing the Hg concentrations. We investigated a potential variation between seasons, and the influence of size, sex and foraging ecology on the individual contamination level.

2. Material and methods

2.1. Study area

The study was conducted in the Nature Reserve "Réserve Naturelle Nationale de Kaw-Roura", French Guiana (4°36′N, 52°07 W) (Fig. 1), a 94.700 ha protected area situated approximately 90 Km southwest of the city of Cayenne. Animals were captured in "Agami Pond", an area situated in the middle of the Nature Reserve, (04°38′N, 52°09′W), a patchwork of herbaceous savannah, swamp forest and open water. Black caimans were sampled during three sampling periods: once during dry season (October 2013), and two samplings during rainy season (May 2014 and May 2015). Caimans were located at night between 19:00pm to 04:00am, using a head lamp, and further captured with a noose.

2.2. Sample collection

A total of 72 individuals were captured, among which 49 adults and subadults were sexed with a ratio of 30 males and 19 females. The body (from the tip of the snout to the cloaca) and total length (including the tail) of each individual were measured with a flexible ruler. We collected a blood sample (~2 mL) through occipital venous sinus puncture, using a syringe with a 30 gauge heparinized needle (heparin sodium). Black caimans were released at the location of their capture directly after biometric measurements and sample collection were performed.

Blood samples were separated as following: 1 mL of each blood sample was centrifugated in order to separate the red blood cells (RBCs) and plasma for isotopic analyses (see below), an additional 1 mL of the total blood was kept in 70% alcohol until further processed at the laboratory for Hg assays. Samples were initially collected to investigate the dietary ecology of Black caimans (Caut et al., 2019). We took the opportunity to further use this sampling set to investigate the Hg concentration in the blood of *Melanosuchus niger*. As a result, the protocol that had been originally applied did not allow us to assess water blood content, and Hg values are therefore presented as dry weight (dw) of the total blood (see below).

2.3. Mercury analysis

One mL of each total blood sample was freeze-dried and grounded to a fine powder. Total Hg concentration (hereafter Hg) in the blood was determined by direct measurement using an atomic absorption spectrometer AMA-254 (Advanced Mercury Analyser-254; Altec®). Analyses were made on at least two replicates of ~3.0 mg dry weight (dw) for each individual. The reproducibility for duplicate samples was approved when Relative Standard Deviation (RSD) was below 10%. The method was validated by the analyses of certified reference material (CRM) produced by the National Research Council of Canada: TORT-2 (Lobster hepatopancreas; certified Hg concentration: 0.27 ± 0.06 μg g^{-1} dw) and TORT-3

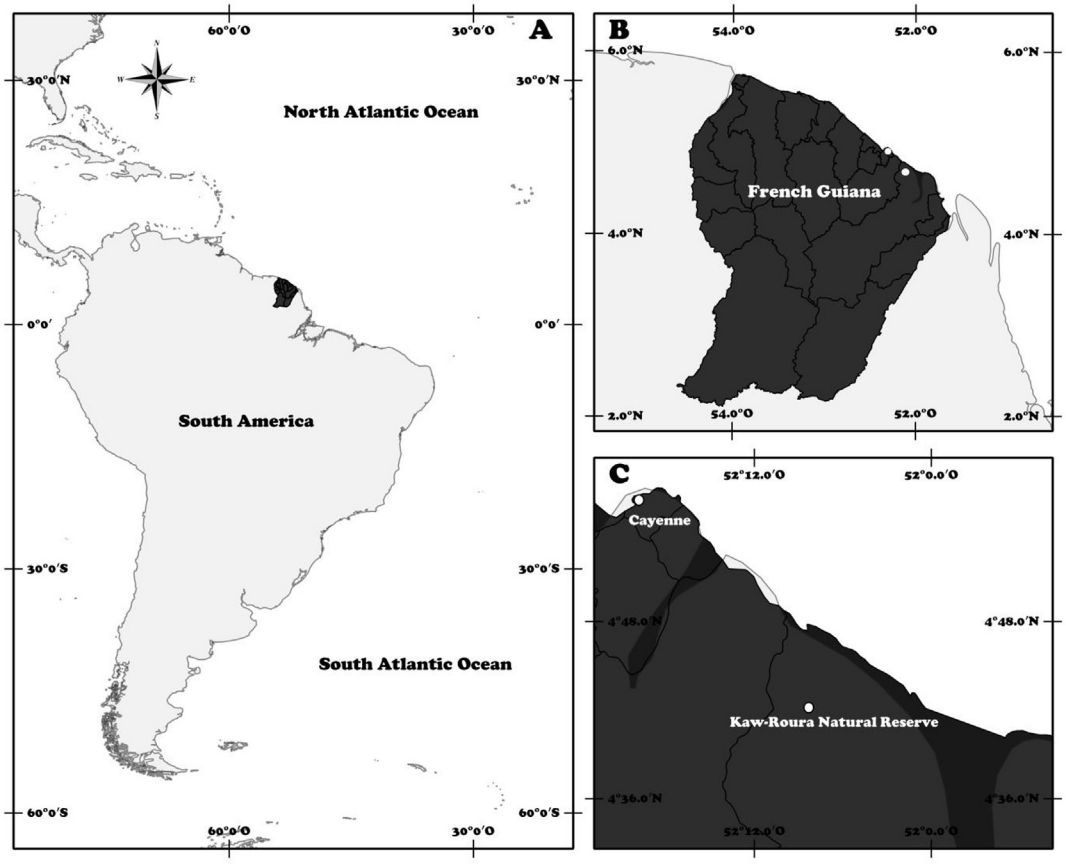


Fig. 1. Geographic location of the study site in South America (A), French Guiana (B), Kaw-Roura Nature Reserve (C).

(Lobster hepatopancreas; certified Hg concentration: 0.29 ± 0.02 μg g^{-1} dw). CRMs were analyzed at the beginning and at the end of the analytical cycle, and between every 10 samples (Chouvelon et al., 2009). Recovery rates of certified reference material were $97.3 \pm 1.0\%$ for TORT-2 ($n = 4$) and $102.0 \pm 1.5\%$ for TORT-3 ($n = 5$). Blanks were included at the beginning of each analytical run and the limit of quantification was 0.05 ng. Hg concentrations in caiman blood are presented in μg.g^{-1} dw.

2.4. Stable isotope analysis

An analysis of nitrogen and carbon stable isotopes was conducted on red blood cells (RBCs) separated from plasma by centrifugation. RBC samples were freeze-dried and then grounded to a fine powder. Aliquots of 0.3–0.4 mg were placed in tin capsules. Stable isotopes were analyzed using a mass spectrometer (IsoPrime 100, Isoprime, UK) associated to a C–N–S elementary analyser (vario MICRO cube, Elementar, Germany). Stable carbon and nitrogen isotope ratios are expressed as ($\delta^{15}N$) or ($\delta^{13}C$) = [($R_{sample}/R_{standard}$)-1]x1000, where R is $^{15}N/^{14}N$ or $^{13}C/^{12}C$ for $\delta^{15}N$ or $\delta^{13}C$. IAEA-CG-6 (−10.4‰) was used as a standard reference for carbon, and IAEA-N1 (+0.4‰) for nitrogen. Ten replicate assays of internal laboratory standards indicated maximum measurement errors (SD) of ±0.2‰ and ±0.15‰ for the nitrogen and carbon isotope measurements, respectively. Further details on isotope analysis are available in Caut et al. (2019).

2.5. Statistical analyses

All analyses were performed using the software R, v.3.2.4 (R development Core Team 2013).

The data was first checked for normality and homogeneity of variances. The relationship between Hg concentration and animal total length was assessed by parametric linear regression. Paired t-tests were used to compare Hg concentrations between the sex, and the season. A linear regression model was used to determine the relationship between Hg concentrations, $\delta^{15}N$ and $\delta^{13}C$ values and to further determine the relationship between caiman length and $\delta^{15}N$ and $\delta^{13}C$ values. The significance for statistical analyses was always set at $p < 0.05$.

3. Results

The Hg concentrations in the 72 sampled individuals ranged from 0.299 to 3.408 μg g^{-1} dw (Table 1). There was a significant, positive relationship between Hg concentration and total body length (Linear regression, $F_{1,70} = 92.37$, $p < 0.0001$, $r^2 = 0.56$, Fig. 2). Adult females and males had a similar size range, respectively 162.9 ± 60.4 cm and 171.0 ± 53.1 cm (Paired *t*-test, $t = 0.48$, $p = 0.64$) and displayed similar Hg concentrations, respectively 1.660 ± 0.694 μg g^{-1} dw and 1.459 ± 0.502 μg g^{-1} dw (Paired *t*-test, $t = 1.09$, $p = 0.28$, Fig. 3).

Seasons (dry and rainy) did not influence Hg concentration (Paired *t*-test, $t = 0.38$, $p = 0.70$), with values of 1.499 ± 0.440 μg g^{-1}

Table 1
Sex, total length (cm) and blood Hg concentration (μg g^{-1} dw) of the Black caiman, *Melanosuchus niger*, from French Guiana.

	N (Males/Females)	Total Length		Hg concentrations	
		Mean ± SD	Min-Max	Mean ± SD	Min-Max
Season					
Dry	18 (11/7)	155.8 ± 35.8	114–278	1.499 ± 0.440	0.741–2.432
Rainy	31 (19/12)	174.9 ± 63.8	95–320	1.559 ± 0.661	0.572–3.408
Sex					
Male	30	171.0 ± 53.1	109–320	1.459 ± 0.502	0.717–2.894
Female	19	162.9 ± 60.4	95–254	1.660 ± 0.694	0.572–3.408

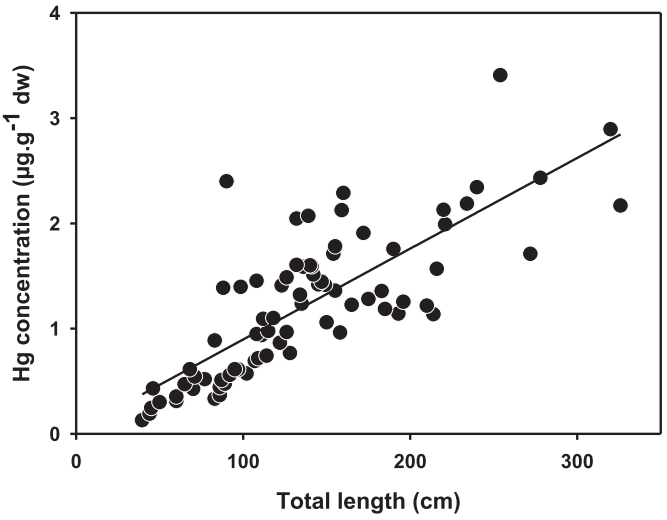


Fig. 2. Relationship between the total length (in cm) and Hg concentration (μg g^{-1} dw) measured in the blood of Black caiman *Melanosuchus niger* from French Guiana (Linear regression, $F_{1,70} = 92.37$, $p < 0.0001$, $r^2 = 0.56$).

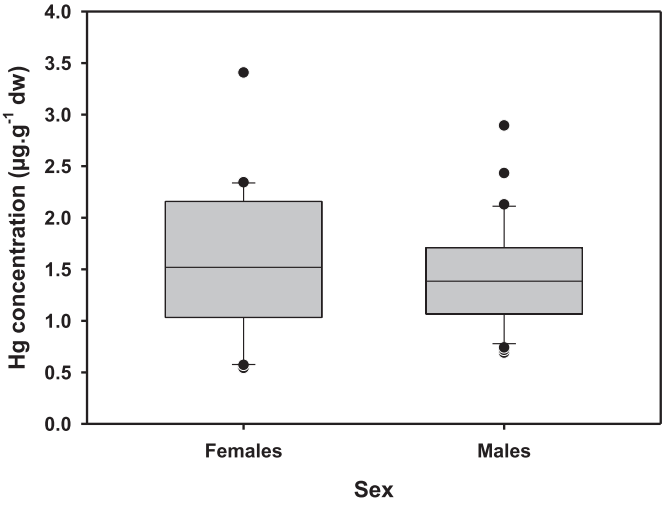


Fig. 3. Hg concentrations in the blood of males and females Black caiman *Melanosuchus niger* from French Guiana. The top and bottom of the boxes represent the first and last quartiles, the line across the box represents the median, the whiskers represent the fifth and ninety-fifth percentiles, and the circles represent outliers.

dw for dry season and 1.559 ± 0.661 μg g^{-1} for rainy season.

The $\delta^{15}N$ was significantly and positively related to caiman total length (linear regression, $F_{1,70} = 68.33$, $p < 0.0001$, $r^2 = 0.49$) and Hg concentration (linear regression, $F_{1,70} = 58.74$, $p < 0.0001$, $r^2 = 0.45$, Fig. 4a). However, the $\delta^{13}C$ was not related to caiman total

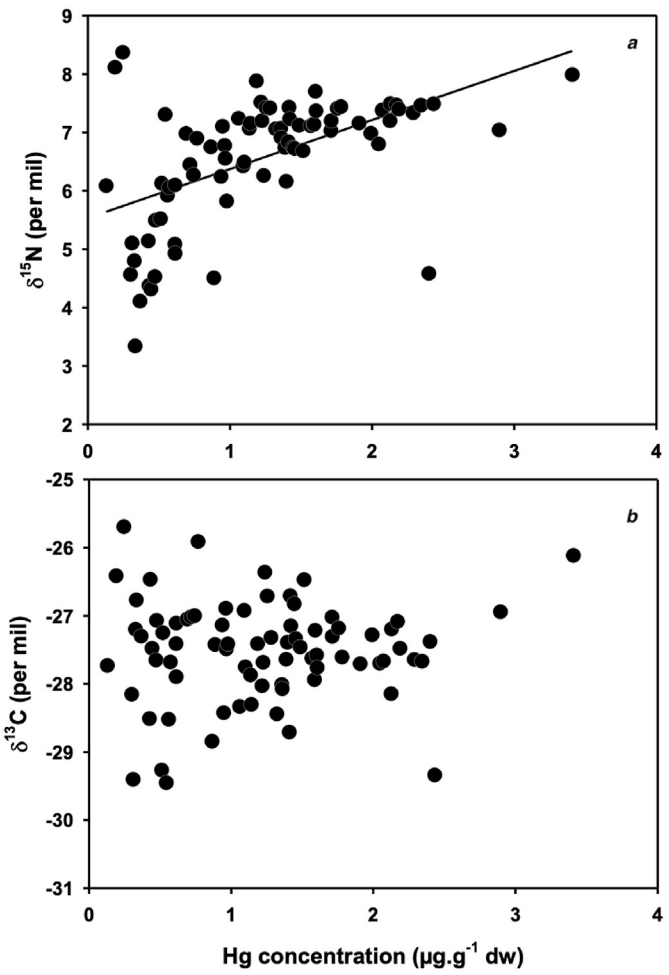


Fig. 4. Relationship between Hg concentration in the blood (μg g^{-1} dw) and stable isotope ratios of (a.) $\delta^{15}N$ (‰) (linear regression, $F_{1,70} = 58.74$, $p < 0.0001$, $r^2 = 0.45$) and (b.) $\delta^{13}C$ (‰) (linear regression, $F_{1,70} = 1.13$, $p = 0.27$, $r^2 = -0.003$) of Black caiman *Melanosuchus niger* from French Guiana.

length (linear regression, $F_{1,70} = 0.86$, $p = 0.36$, $r^2 = -0.002$) nor Hg concentration (linear regression, $F_{1,70} = 1.13$, $p = 0.27$, $r^2 = -0.003$, Fig. 4b).

4. Discussion

Our results show that Black caimans in French Guiana bioaccumulate Hg. As a consequence, Hg concentration increases with body size and is further linked to their trophic position ($\delta^{15}N$). We did not find any sexual or seasonal correlation with the Hg concentration.

Blood is a universally used matrix to measure Hg exposure in a

wide array of organisms (Lommel et al., 1992; Henny et al., 2002; Eggins et al., 2015). Our results show a significant positive relationship between the total length of Black caiman and the Hg concentration in the blood (Fig. 2). Mercury is being transported through the blood to different tissues, such as those involved in detoxification (mainly the liver), storage (muscles), excretion (kidneys) and elimination (keratinized tissues). In reptiles, Hg concentrations of blood are related to Hg concentrations of internal tissues because of the dynamic transfer between these matrices (Burger et al., 2007; Eggins et al., 2015; Nilsen et al., 2017). Therefore, Hg values in the blood reflect an overall Hg concentration in the internal tissues. Our results also confirm the usefulness of blood to determine the Hg contamination in caimans (Eggins et al., 2015; Marrugo-Negrete et al., 2019).

Several studies in crocodilians already reported a linear increase of Hg with age in various tissues (Yanochko et al., 1997; Burger et al., 2000; Rumbold et al., 2002; Schneider et al., 2012). Age and size are generally correlated (i.e. Eaton and Link, 2011), yet, in wild populations, detailed information on age is rarely available. The relationship between crocodilian size and Hg concentration in tissues such as blood, scutes, claws, muscles and liver shows that Hg is bioaccumulated across the life of an individual (Burger et al., 2000; Schneider et al., 2015; Lázaro et al., 2015; Marrugo-Negrete et al., 2019). Although the Hg concentration in various tissues and the body size are positively correlated in some crocodilian species (i.e., *Alligator mississippiensis, Caiman crocodilus, Melanosuchus niger, Caiman yacare*), we emphasize that this pattern has not been detected in other species (i.e., *Crocodylus acutus, Crocodylus moreletii*, Yanochko et al., 1997; Burger et al., 2000; Rainwater et al., 2007; Schneider et al., 2015; Lázaro et al., 2015; Marrugo-Negrete et al., 2019). Such divergent findings may highlight the importance of relatively large sample sizes associated with significant body size ranges in order to robustly assess the relationship between Hg and individual traits. Mercury concentrations are significantly linked to $\delta^{15}N$ values (a proxy used to discriminate the trophic level) (Fig. 4a). Results indicate that the Hg concentration is depending on the ontogenetic change in the trophic position in *M. niger* (Caut et al., 2019). Our results do not show a relationship between Hg concentration and $\delta^{13}C$ (a proxy used to discriminate different types of habitat) for *M. niger* (Fig. 4b). This result shows that the ontogenetic change in the foraging ecology of *M. niger* does not induce a change in foraging habitats in the studied population. We did not find any seasonal influence on Hg concentrations, suggesting that the trophic ecology of *M. niger* at our study site does not significantly vary across seasons.

We did not find any variation in Hg concentrations between males and females (Table 1, Fig. 3). A possible mechanism of Hg elimination in females is the maternal transfer of Hg to the eggs: Females use their energy storage (e.g. body fat and proteins) during vitellogenesis, which may induce a transfer of the Hg stored in their tissues towards their eggs (Nilsen et al., 2020) as reported for birds for instance (e.g., Lewis et al., 1993). This process would lead to lower concentrations in the blood of females that recently laid eggs, compared to males. Therefore, our results suggest that either 1), Hg remobilization did not occur, or more likely, 2) this process is not significant enough to be detected in the blood of female Black caimans. Future studies should investigate the level of Hg transfer to the eggs during vitellogenesis in female *M. niger*, as well as in other crocodilian species.

It is important to emphasize that the relatively low turn-over rates of erythrocytes in crocodilians may have obscured putative influences of sex and season on Hg levels. Indeed, blood is known to reflect short-time Hg exposure in birds or mammals (Bearhop et al., 2000). For instance, the lifetime of erythrocytes is up to two months in birds and up to four months in mammals (reviewed in Rodnan et al., 1957; Monteiro and Furness, 2001). In crocodilians, the lifetime of erythrocytes can last up to 3 years (Cline and Waldmann, 1962). Future studies are required to assess whether low turnover rates of erythrocytes in crocodilians influence short-scale, temporal variations in Hg concentrations.

Mercury concentrations in the blood of *M. niger* in French Guiana (1.284 ± 0.672 μg g^{-1} dw) are higher than in other South American species with a maximum value of 0.325 ± 0.105 μg g^{-1} dw (Table 2; Marugo-Negrete et al., 2019). However, limited data is available from this geographic area (Table 2). The values we report in the blood of *M. niger* are similar to what has been determinated in the muscle of the same species in Brazil (Table 2). This is especially interesting as Hg concentrations in the muscle usually tends to be higher than in the blood (Schneider et al., 2012). Insufficient data on Hg in crocodilians from the Americas make comparison difficult, except for the United States where the American alligator is well documented (Table 2). Clearly, future studies are required in order to provide a complete background to perform substantial comparisons. In addition, the use of other tissues where sampling is less invasive (i.e.., claws and scutes, Lázaro *al*., 2015; Marrugo-Negrete et al., 2019), should be considered, for it is already a conventional method in other reptile species (e.g., Slimani et al., 2018; Lemaire et al., 2018; Beau et al., 2019).

Table 2
Review of Hg concentration (μg g^{-1} dw) reported in crocodilians. TL stands for Total Length and SVL for Snout-Vent-Length (cm). ([a] Original data reported in wet weight, transformed in dry weight with a factor 3.8 calculated by Jeffree et al., 2001 for muscles;[b] original data reported in wet weight, transformed in dry weight using a factor 5).

Species	Location	Tissue	n	Length (cm)		Hg (μg g^{-1} dw)		Reference
				Mean ± SD	Min-Max	Mean ± SD	Min-Max	
Black caiman *Melanosuchus niger*	Rio Purus, Brazil	Muscles	11	107.5 ± 31.4 (SVL)	75.3–190.9 (SVL)	1.93[a]	0.69–4.06[a]	Schneider et al. (2012)
Black caiman *Melanosuchus niger*	Rio Purus, Brazil	Muscles	16	102 ± 27 (SVL)	75–191 (SVL)	0.669 ± 0.369[a]	–	Eggins et al. (2015)
Black caiman *Melanosuchus niger*	Mamirauá Reservoir, Brazil	Muscles	60	107–309 (TL)	107–309 (TL)	1.457 ± 0.433[a]	–	Correia et al. (2014)
American Alligator *Alligator mississippiensis*	South Carolina	Total Blood	–	–	–	2.19 ± 0.38	–	Jagoe et al. (1998)
American Alligator *Alligator mississippiensis*	Florida	Total Blood	37	92.1 ± 31.6 (SVL)	43.9–153.5 (SVL)	0.965[b]	0.280–6.900[b]	Nilsen et al. (2017)
Spectacled Caiman *Caiman crocodilus*	La Mojana, Colombia	Total Blood	22	57.2 ± 3.5 (TL)	–	0.325 ± 0.105[b]	–	Marrugo-Negrete et al. (2019)
Spectacled Caiman *Caiman crocodilus*	La Mojana, Colombia	Total Blood	23	57.5 ± 6.8 (TL)	–	0.07 ± 0.04[b]	–	Marrugo-Negrete et al. (2019)
Black caiman *Melanosuchus niger*	Kaw, French Guiana	Total Blood	72	143.2 ± 61.3 (TL)	46–326 (TL)	1.284 ± 0.672	0.30–3.41	Our study

5. Conclusion

Overall, the use of blood of *M. niger* is informative regarding the Hg concentration and it extends the use of Crocodilians to monitor the environmental Hg contamination. To do so, precise individual information (e.g., size and diet or trophic position) is required. For instance, the change of the trophic position has a significant impact on the level of Hg contamination between juveniles (feeding on low trophic level prey) and adults (feeding mostly on high trophic level prey). Therefore, it is essential to take their diet into account to compare levels of Hg contamination between different sites or populations. Additionally, future studies are required to assess whether the concentrations of Hg we have found in *M. niger* pose a threat to the species.

CRediT authorship contribution statement

Jérémy Lemaire: Conceptualization, Formal analysis, Software, Funding acquisition, Writing - original draft, Writing - review & editing. **Paco Bustamante:** Conceptualization, Writing - original draft, Writing - review & editing, Supervision. **Olivier Marquis:** Conceptualization, Investigation, Writing - original draft, Funding acquisition, Supervision. **Stéphane Caut:** Conceptualization, Investigation, Funding acquisition, Writing - original draft. **François Brischoux:** Conceptualization, Writing - original draft, Writing - review & editing, Supervision.

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.

Acknowledgments

We are grateful to the authorities of Conseil Scientifique Régional du Patrimoine Naturel (CSRPN) and La Direction de l'Environnement, de l'Aménagement et du Logement de Guyane (DEAL), who authorized our fieldwork. We would also like to thank M. Bacques, V. François, D. Guiral, G. Lepoint, N. Sturaro, M. Sarrazin and S. Charles for their help in the field as well as E. Angulo, MJ. Jowers, I. Gomez Muestre, JP. Macé, and C. Franklin for other assistance during this project. We are grateful to A. Smith from Cincinnati Zoo and Botanical Garden, and R. Mangione for the English revision. This work was supported by the Office de l'Eau de Guyane; the Agence Française pour la Biodiversité; the Direction de l'Environnement, de l'Aménagement et du Logement de Guyane; Zoo de Paris; Fondation d'entreprise Hermès; Consejo Superior de Investigaciones Cientıficas (CSIC) research contracts to SC and the Estacion Biologica de Donana; Fonds de Dotation pour la Biodiversity and the private company Lacoste; the National Geographic Society/Waitt Grants Program (GRANT #W269-13) and the CNRS. We are also very grateful to C. Churlaud and M. Brault-Favrou from the plateforme Analyses Elémentaires of the LIENSs laboratory for their advices during Hg analyses. The CPER (Contrat de Projet Etat-Région) and the FEDER (European regional Development Fund) are acknowledged for funding the AMA. The Institut Universitaire de France (IUF) is acknowledged for its support to P. Bustamante as a Senior Member.

References

Atwell, L., Hobson, K.A., Welch, H.E., 1998. Biomagnification and bioaccumulation of mercury in an artic marine food web: insights from stable nitrogen isotope analysis. Can. J. Fish. Aquat. Sci. 55 (5), 1114–1121.

Bearhop, S., Ruxton, G.D., Furness, R.W., 2000. Dynamics of mercury in blood and feathers of great-skuas. Environ. Toxicol. Chem. 19, 1638–1643.

Beau, F., Bustamante, P., Michaud, B., Brischoux, F., 2019. Environmental causes and reproductive correlates of mercury contamination in European pond turtles (*Emys orbicularis*). Environ. Res. 172, 338–344.

Benoit, J.M., Gilmour, C.C., Heyes, A., Mason, R.P., Miller, C.L., 2003. Geochemical and biological controls over methylmercury production and degradation in aquatic ecosystems. Am. Chem. Soc. Symp. Ser. 835, 262–297.

Bontemps, D.R., Cuevas, E., Ortiz, E., Wunderle Jr., J.M., Joglar, D.R., 2016. Diet of the non-native spectacled caiman (*Caiman crocodilus*) in Puerto Rico. Manag. Biol. Invasion. 7, 287–296.

Burger, J., Campbell, K.R., Murray, S., Campbell, T.S., Gaines, K.F., Jeitner, C., Shukla, T., Burke, S., Gochfeld, M., 2007. Metals levels in blood, muscle and liver of water snakes (*Nerodia* spp.) from New Jersey, Tennessee and South Carolina. Sci. Total Environ. 373, 556–563.

Burger, J., Gochfeld, M., Rooney, A.A., Orlando, E.F., Woodward, A.R., Guillette Jr., L.J., 2000. Metals and metalloids in tissues of American alligators in three Florida lakes. Arch. Environ. Contam. Toxicol. 38, 501–508.

Camus, A.C., Mitchell, M.M., Williams, J.F., Jowett, P.L.H., 1998. Elevated lead levels in farmed American alligators *Alligator mississipiensis* consuming nutria *Myocastor coypus* meat contaminated by lead bullets. J. World Aquacult. Soc. 3, 370–376.

Caut, S., Francois, V., Bacques, M., Guiral, D., Lemaire, J., Lepoint, G., Marquis, O., Sturaro, N., 2019. The dark side of the black caiman: shedding light on species dietary ecology and movement in Agami Pond, French Guiana. PloS One 14 (6), e0217239.

Chouvelon, T., Warnau, M., Churlaud, C., Bustamante, P., 2009. Hg concentrations and related risk assessment in coral reef crustaceans, molluscs and fish from New Caledonia. Environ. Pollut. 157, 331–340.

Cline, M., Waldmann, T.A., 1962. Effect of temperature on red cell survival in the alligator. Proc. Soc. Exp. Biol. Med. 111, 716–718.

Compeau, G.C., Bartha, R., 1985. Sulphate reducing bacteria: principal methylators of mercury in anoxic estuarine sediment. Appl. Environ. Microbiol. 50, 498–502.

Cook, R.A., Behler, J., Braeaitis, P., 1991. Elevated heavy metal concentrations in captive crocodilians-two cases. In: Procceedings of 1989 Annual Meeting of American Association of Zoo Veterinarians, p. 151. Greensboro, North Carolina.

Correia, J., Cesar, R., Marsico, E., Diniz, G.T., Zorro, M.C., Castilhos, Z., 2014. Mercury contamination in alligators (*Melanosuchus niger*) from Mamiraúá Reservoir (Brazilian Amazon) and human health risk assessment. Environ. Sci. Pollut. Res. 21, 13522–13527.

Day, R.D., Christopher, S.J., Becker, P.R., Whitaker, D.W., 2005. Monitoring mercury in loggerhead sea turtle, *Caretta caretta*. Environ. Sci. Technol. 39, 437–446.

De Thoisy, B., Hrbek, T., Farias, I.P., Vasconcelos, W.R., Lavergne, A., 2006. Genetic structure, population dynamics, and conservation of black caiman (*Melanosuchus niger*). Biol. Conserv. 133 (4), 474–482.

De Lacerda, L.D., 2003. Updating global Hg emissions from small-scale gold mining and assessing it environmental impacts. Environ. Geol. (Berl.) 43, 308–314.

Eaton, M.J., Link, W.A., 2011. Estimating age from recapture data: integrating incremental growth measures with ancillary data to infer age-at-length. Ecol. Appl. 21 (7), 2487–2497.

Eggins, S., Schneider, L., Krikowa, F., Vogt, R.C., Silveira, R.D., Maher, W., 2015. Mercury concentrations in different tissues of turtle and caiman species from Rio Purus, Amazonas, Brazil. Environ. Toxicol. Chem. 34 (12), 2771–2781.

Ericksen, J.A., Gustin, M.S., Schorran, D.E., Johnson, D.W., Lindberg, S.E., Coleman, J.S., 2003. Accumulation of atmospheric mercury in forest foliage. Atmos. Environ. 37, 1613–1622.

Fort, J., Robertson, G.J., Grémillet, D., Traisnel, G., Bustamante, P., 2014. Spatial ecotoxicology: migratory arctic seabirds are exposed to mercury contamination while overwintering in the northwest atlantic. Environ. Sci. Technol. 48 (19), 11560–11567.

Fréry, N., Maury-Brachet, R., Maillot, E., Deheeger, M., de Mérona, B., Boudou, A., 2001. Gold-Mining activities and mercury contamination of Native Amerindian communities in French Guiana: key role of fish in dietary uptake. Environ. Health Perspect. 109, 449–456.

Fujisaki, I., Hart, K.M., Mazzotti, F.J., Cherkiss, M.S., Sartain, A.R., Jeffery, B.M., Beauchamp, J.S., Denton, M., 2014. Home range and movements of American alligator (*Alligator mississippiensis*) in an estuary habitat. Anim. Biotelemetry 2, 8.

Henny, C., Hill, E., Hoffman, D., Spalding, M., Grove, R., 2002. Nineteenth century mercury: hazard to wading birds and cormorants of the Carson River, Nevada. Ecotoxicology 11, 213–231.

Hsu-Kim, H., Eckley, C.S., Achá, D., Feng, X., Gilmour, C.C., Jonsson, S., Mitchell, C.P., 2018. Challenges and opportunities for managing aquatic mercury pollution in altered landscapes. Ambio 47 (2), 141–169.

Hutton, J., 1989. Movement, home range, dispersal and the separation of size classes in nile crocodiles. Am. Zool. 29, 1033–1049.

Jagoe, C.H., Arnold-Hill, B., Yanochko, G.M., Winger, P.V., Brisbin JR., I.L., 1998. Mercury in alligators (*Alligator mississipiensis*) in the southeastern United States. Sci. Total Environ. 213, 255–262.

Jeffree, R.A., Markich, S.J., Twining, J.R., 2001. Element concentrations in the flesh and osteoderms of estuarine crocodiles (*Crocodylus porosus*) from the Alligator rivers region, northern Australia: biotic and geographic effects. Arch. Environ. Contam. Toxicol. 40, 236–245.

Lázaro, W.L., de Oliveira, R.F., dos Santos-Filho, M., da Silva, C.J., Malm, O., Ignácio, Á.R.A., Díez, S., 2015. Non-lethal sampling for mercury evaluation in

crocodilians. Chemosphere 138, 25–32.
Lemaire, J., Bustamante, P., Olivier, A., Lourdais, O., Michaud, B., Boissinot, A., Galán, P., Brischoux, F., 2018. Determinants of mercury contamination in viperine snakes, *Natrix maura*, in Western Europe. Sci. Total Environ. 635, 20–25.
Lewis, S.A., Becker, P.H., Furness, R.W., 1993. Mercury levels in eggs, tissues, and feathers of herring gulls *Larus argentatus* from the German Wadden Sea Coast. Environ. Pollut. 80, 293–299.
Lommel, A., Kruse, H., Müller, E., Wassermann, O., 1992. Organochlorine pesticides, octachlorostyrene, and mercury in the blood of Elb River residents, Germany. Arch. Environ. Contam. Toxicol. 22–1, 14–20.
Magnusson, E.W., Albertina, P.L., 1991. The ecology of a cryptic predator, *Paleosuchus trigonatus*, in a tropical rainforest. J. Herpetol. 25 (1), 41–48.
Marrugo-Negrete, J., Durango-Hernández, J., Calao-Ramos, C., Urango-Cárdenas, I., Díez, S., 2019. Mercury levels and genotoxic effect in caimans from tropical ecosystems impacted by mining. Sci. Total Environ. 664, 899–907.
Mason, R.P., Reinfelder, J.R., Morel, F.M.M., 1995. Bioaccumulation of Mercury and Methylmercury. Mercury as a Global Pollutant, pp. 915–921.
Minagawa, M., Wada, E., 1984. Stepwise enrichment of ^{15}N along food chains: further evidence and the relation between $\partial^{15}N$ and animal age. Geochem. Cosmochim. Acta 48, 1135–1140.
Monteiro, L.R., Furness, R.W., 2001. Kinetics, dose-response, and excretion of methylmercury in free-living cory's shearwaters. Environ. Sci. Technol. 35, 739–746.
Nilsen, F.M., Kassim, B.L., Delaney, J.P., Lange, T.R., Brunell, A.M., Guillette Jr., L.J., Long, S.E., Schock, T.B., 2017. Trace element biodistribution in the American alligator (*Alligator mississippiensis*). Chemosphere 181, 343–351.
Nilsen, F.M., Rainwater, T.R., Wilkinson, P.M., Brunell, A.M., Lowers, R.H., Bowden, J.A., Guillette, L.J., Long, S.E., Schock, T.B., 2020. Examining maternal and environmental transfer of mercury into American alligator eggs. Ecotoxicol. Environ. Saf. 189, 110057.
Ottenbros, I.B., Boerleider, R.Z., Jubitana, B., Roelleveld, N., Scheepers, P.T.J., 2019. Knowledge and awareness of health effects related to the use of mercury in artisanal and small-scale gold mining in Suriname. Environ. Int. 122, 142–150.
Pinnegar, J., Polunin, N., 2000. Contribution of stable-isotope data to elucidating food webs of Mediterranean rocky littoral fishes. Oecologia 122, 399–409.
Post, D.M., 2002. Using stable isotopes to estimate trophic position: models, methods, and assumptions. Ecology 83, 703–718.
Power, M., Klein, G.M., Guiguer, K.R.R.A., Kwan, M.K.H., 2002. Mercury accumulation in the fish community of a sub-Artic lake in relation to trophic position and carbon sources. J. Appl. Ecol. 39 (5), 819–830.
R Core Team, 2013. R: A Language and Environment for Statistical Computing. R foundation for Statistical Computing, Vienna, Austria. http://www.R-project.org/.
Radloff, G.G.T., Hobson, K.A., Leslie, A.J., 2012. Characterising ontogenetic niche shifts in Nile crocodile using stable isotope ($\partial^{13}C$, $\partial^{15}N$) analyses of scute keratin. Isot. Environ. Health Stud. 48, 239–456.
Rainwater, T.R., Wu, T.H., Finger, A.G., Cañas, J.E., Yu, L., Reynolds, K.D., Coimbatore, G., Barr, B., Platt, S.G., Cobb, G.P., Anderson, T.A., McMurry, S.T., 2007. Metals and organochlorine pesticides in caudal scutes of crocodiles from Belize and Costa Rica. Sci. Total Environ. 373, 146–156.
Rocha, R., Olivero, V., Caballero, G., 2018. Impact of fold mining associated with mercury contamination in surface soil of San Martin de Loba, south of Bolivar (Colombia). Rev. Int. Contam. Ambient. 34 (1), 93–102.
Rodnan, G., Ebaugh, F.G., Fox, M.R.S., Chambers, D.M., 1957. The life span of the red blood cell and the red blood cell volume in the chicken, pigeon and duck as estimated by the use of Na_2, $Cr^{51}O_4$: with observations on red cell turnover rate in the mammal, bird and reptile. Blood 12, 355–366.
Rumbold, D.G., Fink, L.E., Laine, K.A., Niemczyk, S.L., Chandrasekhar, T., Wankel, S.D., Kendall, C., 2002. Levels of mercury in alligators (*Alligator mississipiensis*) collected along a transect trough the Florida Everglades. Sci. Total Environ. 297, 239–252.
Scheuhammer, A.M., Sandheinrich, M.B., 2007. Recent advances in the toxicology of methylmercury in wildlife. Ecotoxicology 17 (2), 67–68.
Schneider, L., Eggins, S., Maher, W., Vogt, R.C., Krikowa, F., Kinsley, L., Eggins, S.M., Da Silveira, R., 2015. An evaluation of the use of reptile dermal scutes as a non-invasive method to monitor mercury concentrations in the environment. Chemosphere 119, 163–170.
Schneider, L., Peleja, R.P., Kluczkovski Jr., A., Freire, G.M., Marioni, B., Vogt, R.C., Da Silveira, R., 2012. Mercury concentration in the spectacled caiman and black caiman (Alligatoridae) of the Amazon: implications for human health. Arch. Environ. Contam. Toxicol. 63, 270–279.
Slimani, T., El Hassani, M.S., Bonnet, M., Bustamante, P., Brischoux, F., Brault-Favrou, M., Bonnet, X., 2018. Large-scale geographic patterns of mercury contamination in Morocco revealed by freshwater turtles. Environ. Sci. Pollut. Res. 25 (3), 2350–2360.
Twining, J.R., Markich, S.J., Prince, K.E., Jeffree, R.A., 1999. Osteoderms of estuarine crocodiles record their enhanced Pb exposure in Kakadu National Park. Environ. Sci. Technol. 33, 4396–4400.
Vanderklift, A., Ponsard, S., 2003. Sources of variation in consumer-diet ∂15N enrichments: a meta-analysis. Oecologia 136, 169–182.
Yanochko, G., Jagoe, C., Brisbin Jr., L., 1997. Tissue mercury concentrations in alligators (*Alligator mississipiensis*) from the Florida Everglades and the Savannah River site, South Carolina. Arch. Environ. Contam. Toxicol. 32, 323–328.