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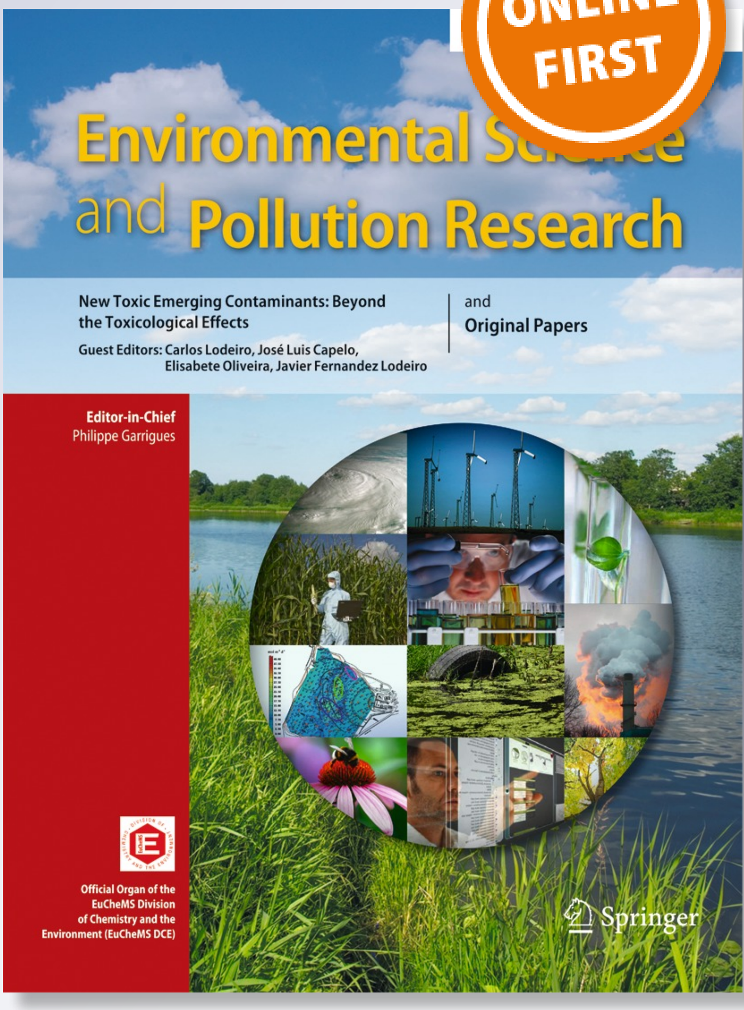

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MULTI-STRESSORS IN FRESHWATER AND TRANSITIONAL ENVIRONMENTS: FROM LEGACY POLLUTANTS TO EMERGING ONES

Mercury contamination levels in the bioindicator piscivorous fish *Hoplias aïmara* in French Guiana rivers: mapping for risk assessment

Régine Maury-Brachet[1] · Sophie Gentes[1] · Emilie P. Dassié[1] · Agnès Feurtet-Mazel[1] · Régis Vigouroux[2] · Valérie Laperche[3] · Patrice Gonzalez[1] · Vincent Hanquiez[1] · Nathalie Mesmer-Dudons[1] · Gilles Durrieu[4] · Alexia Legeay[1]

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Abstract

In French Guiana, native populations present high level of mercury contamination, which has been linked to the consumption of contaminated fishes. The goal of this study is to undertake a cartography of mercury contamination levels in fishes from the six main Guiana rivers. The selected species for this study is the ubiquitous piscivorous fish *Hoplias aimara*. A total number of 575 fishes from 134 discrete fishing sites are regrouped into 51 river sectors. Results from this study permits to rank the six main Guiana rivers by their mean level of contamination: Oyapock (0.548 mg kg^{-1}), Comté (0.624 mg kg^{-1}), Maroni (0.671 mg kg^{-1}), Approuague (0.684 mg kg^{-1}), Mana (0.675 mg kg^{-1}), and Sinnamary (1.025 mg kg^{-1}). The contamination is however not spatially homogenous along each river, and a map of the different levels of mercury contamination in fishes is provided. Sectors of low mean Hg contamination are observed both upstream (0.471 mg kg^{-1}) and downstream (0.424 mg kg^{-1}), corresponding to areas without any influence of gold mining activities and areas under the influence of estuarine dilution, respectively. Anoxia and gold mining activities are found to be the two main factors responsible for the high mercury concentration in fish muscles. While mean levels of mercury contaminations are higher in anoxia areas (1.029 mg kg^{-1}), contaminations induced by gold mining activities (0.717 mg kg^{-1}) present the most harmful consequences to human populations. No significant differences in Hg concentrations are observed between 2005 and 2014 for neither a pristine nor a gold mining area, while Hg concentration differences are observed between former (0.550 mg kg^{-1}) and current gold mining sites (0.717 mg kg^{-1}).

Keywords Mercury · Piscivorous fish · Freshwater · Gold mining · Hydropower · Swamp

Introduction

Mercury (Hg) is a chemical element that occurs naturally in the environment through volcanism, soil erosion, and ocean degassing. Human activity is also a major actor in releasing Hg into the environment via power plants, incinerators, industrial activities, and gold mining activities (Streets et al. 2009; Driscoll et al. 2013; Kocman et al. 2017). Mercury is classified as a priority pollutant by the World Health Organization (WHO 1990) and the European Water Framework Directive (WFD 2013/39/EC). The toxicity of Hg is speciation dependent. There are three forms of Hg: (i) the elementary mercury (Hg°) presents under liquid and volatile forms, (ii) the oxidized divalent mercury (Hg^{2+}) presents under free or complexed forms, and (iii) the methylmercury (CH_3Hg^+), also called monomethylmercury (MeHg). MeHg is the most toxic form for living organisms. Under anoxic and suboxic conditions, microorganisms transform inorganic mercury into MeHg (Watras et al. 1998; Wiener et al. 2002; Winch et al. 2009). In the environment, this process happens principally at the interface "water sediment" (King et al. 2000) and inside

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✉ Régine Maury-Brachet
regine.maury-brachet@u-bordeaux.fr

[1] Université de Bordeaux, UMR EPOC 5805, Station Marine, Place du Docteur Peyneau, 33120 Arcachon, France

[2] Hydreco, Kourou, Guyane, France

[3] French Geological Survey, Laboratoire d'Environnement et d'Écotechnologies, Orléans, France

[4] Université de Bretagne Sud, LMBA UMR 6205, Lorient, France

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the biofilm (Acha et al. 2005; Gentès et al. 2013). Chemical characteristics of MeHg (remanence, high diffusive capacity through cellular membranes, and high half-life) give it a high capacity of bioaccumulation into organisms as well as biomagnification along the foodweb (Watras et al. 1998), mainly through trophic pathway (Harris and Bodaly 1998). MeHg concentrations could start as low as ng l^{-1} in river's water and could go to mg kg^{-1} into piscivorous fishes' muscles (Coquery et al. 2003; Maury-Brachet et al. 2006; Castilhos et al. 2015; Bastos et al. 2016; Bradley et al. 2017).

French Guiana is widely impacted by mercury contaminations mainly because of gold mining activities present all over the territory. These activities started in the 1850s and followed gold prices, with a decrease in the 1920, and a constant increase from the 1980s to today. Since the 1850s, it has been estimated that more than 300 t of Hg has been released into French Guiana gold-bearing soils (Picot et al. 1993; Bizi et al. 2005; Cottard and Laperche 2012; Laperche et al. 2014). Since 2006, the utilization of Hg is prohibited in French Guiana; therefore, legal gold mining sites stopped using it. Illegal gold mining activity, which represents more than 98% (PAG 2017) of the total gold mining activities in French Guiana, continues to release Hg in the environment. This illegal activity, more than having disastrous impact on the environment, is also directly harmful for native populations. A high exposure to Hg leads to serious health issues (Grandjean et al. 1994; Mergler and Lebel 2001; Mergler et al. 2007). In French Guiana, studies have shown that native populations present high level of Hg contaminations (Cordier et al. 1997; Fréry et al. 2001; Cardoso et al. 2010; Fujimura et al. 2011) due to the consumption of contaminated fishes (Harada 1995; Nielsen et al. 1997; Myers et al. 2003). Amerindian populations such as the Wayanas, Teko, and Wayampi are settled, and their diet is mainly composed of fishes caught locally (Fréry et al. 1999). Illegal gold mining activities have also strong impacts on the fishes' health, biomass, and diversity. Soil leaching and deforestation due to gold mining activities increase the water turbidity which creates hostile living habitats for fishes and therefore decreases fishes' biomass and biodiversity (Brosse et al. 2011; Costa et al. 2012; Tudesque et al. 2012; Laperche et al. 2014; Dezécache et al. 2017). Additionally, illegal gold miners are settling near the rivers and therefore tend to reduce the quantity of fishes (Castilhos et al. 2015; Mol et al. 2001; Thomassin et al. 2017). For the remaining of the manuscript, references to gold mining activities mean, indeed, illegal ones.

It is of environmental and societal importance to determine the spatial distribution of Hg contamination levels in fishes from the French Guiana territory. This will be helpful to the French Guiana community as well as to policyholders to pinpoint contaminated vs pristine fishing areas. A bioindicator fish, *Hoplias aimara*, is used to determine Hg concentrations from various sites along the six main French Guiana rivers. Objectives of this study are to (1) obtain a spatial repartition of fish Hg contaminations from the six main Guiana rivers, (2) perform an inter-river Hg level comparison and establish possible causes of contamination, and (3) evaluate the temporal Hg contamination evolution from two times period (2005 and 2014) at a pristine and a gold mining–impacted site.

Material and methods

The bioindicator species: *Hoplias aimara*

The targeted fish is *Hoplias aimara*. This piscivorous fish belongs to the Characiforms order and Erytrinidae family. It is a high trophic-level species, well represented in the six main watersheds in French Guiana. This species has a sedentary life; it only does limited displacements during the wet season, to nearby creeks to catch prey (Planquette et al. 1996). This fish is therefore representative of the capturing site's environmental conditions. *Hoplias aimara* is also one of the most fished species by the native population for consumption, which allows the assessment of the human risk stem from consuming these fishes. All these characteristics make this species an excellent bioindicator of mercury contamination.

Sampling strategy

French Guiana is located in the northern part of South America within 2–6° N and 51° 30′–54° 30′ W. It is surrounded by the Surinam to the west, Brazil to the east and south, and the Atlantic Ocean to the north (Fig. 1). French Guiana has an important hydrographic system connected to six main rivers: Maroni, Mana, Sinnamary Comté, Approuague, and Oyapock, from west to east. A total number of 134 discrete fishing sites along these six rivers are selected. These sites are regrouped into 51 sectors in order to (i) obtain enough individuals per sectors, (ii) average sites of similar fish-living conditions (territory of about 10 km), and (iii) recover sites that are under similar levels of contamination.

Fishing campaigns, from two distinct programs, took place concurrently from 2003 to 2006: (1) a program ran by the French National Research Institute (Centre National de la Recherche Scientifique (CNRS)) and (2) a program ran by a French Geological Survey Institute. The first program was based on voluntary work from local populations. More than 100 sampling kits were handed out to volunteers (schools, free clinics, police stations, fishermen, and local researchers) with the necessary tools to identify and describe fish species, dissect muscle, and store samples (4% formalin). Each sampling kit included a waterproof barrel, two measuring tapes, one scalpel with three blades, two pliers, two pencils, 20 water proof small bottles filled with deionized water and formalin at 4%, 20 plastic labels, and 2 markers. An explanatory

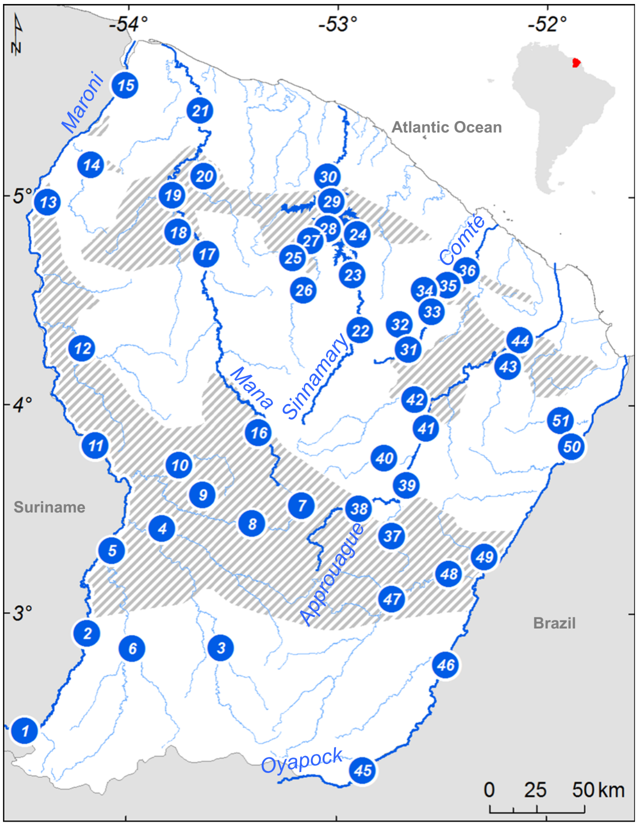

Fig. 1 Location of the 51 sectors on the six French Guiana rivers: Maroni, Mana, Sinnamary, Approuague, and Oyapock. Gray hatched areas correspond to areas under current gold mining activities

document described how to recognize the wanted fish species and how to dissect fish muscles. On a provided index card, volunteers had to indicate the fishing site, fishing date, his/her name, and the standard fish length (length from the snout to the base of the tail). Additionally to volunteer workers, French scientists organized four fishing campaigns during which they recorded similar information that the ones requested from the volunteers. During the first campaign, numerical shoots of the fishes were taken to build the fish recognition guide that was included in the volunteer's kit. Fish species determination was possible thanks to already published studies of Guiana fishes (Boujard et al. 1997; Keith et al. 2000; Kullander and Nijssen 1989; Le Bail et al. 2000; Planquette et al. 1996). The second program realized that 12 campaigns were both fishes and sediments were sampled. These campaigns targeted inhabited areas.

The local population uses fishing traps (Dundee Shop—Cayenne) made of a large self-shoeing hook at the end of a steel rope. During the fishing campaigns, scientists used both fishing traps and 50-m-long trammel fishing nets. A trammel is a fishing net composed of three layers of netting. Trammels for this study were made in continuous nylon and had a central layer with a 35-mm mesh sandwiched between two taut outer layers of 200 mesh (Etablissement Mondiet—Gironde). Trammels were kept vertical by floats located on the top net layer and weights on the bottom net layer.

A total number of 721 fishes were collected: 480 fishes from the CNRS program and 241 fishes from the French Geological Survey program (Laperche et al. 2007, 2014).

In addition to the data obtained by these two campaigns, we received data from another French program (RIMNES project: Mercury isotope fractionation and NOTCH/apoptosis biomarkers: new tracers linking Environment and Health) that ran between 2012 and 2014. Eighteen fishes were sampled during two campaigns that targeted the upstream section of the Oyapock River, in sectors "Trois Sauts" (45) and "Saut Camopi" (48). These data will allow an estimation of the temporal evolution of fish contamination.

Data processing

Standard lengths of the 721 fishes range from 10 to 110 cm (Fig. 2). The selected species for this study, *Hoplias aimara*, looks morphologically similar to *Hoplias malabaricus*. These two species present, however, different biometric characteristics. *H. malabaricus* maximum length is 38 cm for 1.8 kg fresh weight (Planquette et al. 1996), while *H. aimara* can reach 120 cm in length for a total fresh weight of 40 kg. Hg concentration in piscivorous fishes is highly dependent of their exposition time to the contamination and therefore highly dependent of their lifespan (Durrieu et al. 2005; Lavigne et al. 2010; and Lucotte et al. 2016). To our knowledge, there is no study on age determination in neither *Hoplias aimara* nor *Hoplias malabaricus*. To exclude all the *H. malabaricus* individuals, only fish with standard length above 40 cm are included in this study. There is a non-linear relationship between length and Hg concentration in piscivorous fishes (Durrieu

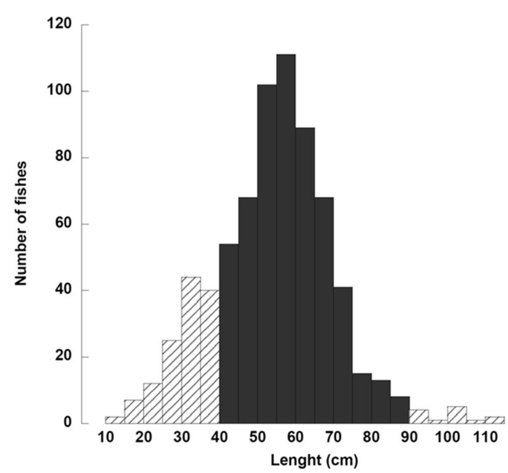

Fig. 2 Distribution of the length of sampled fishes. Fishes of size inferior to 40 cm were casted off as they might include individuals from two different species. Fishes of size superior to 90 cm were removed; they can be considered as outliers (they represent the 2% of the end tail of the distribution)

et al. 2005; Lavigne et al. 2010; Lucotte et al. 2016). It is therefore important to compare individuals from a narrow range of length (Lavigne et al. 2010). Clustering individuals by narrow ranges of length decrease drastically the number of fishes per site, and do not allow inter-site comparisons. We therefore decided to remove fishes whose length is located in the 5% of the end tail distribution. This study focus on 575 fishes from 40 to 90-cm length (Fig. 2).

Mercury analyses

For each fish, an aliquot of muscle is sampled and dried at 45 °C in oven for 48 h. Total Hg concentrations in fish muscles are determined by flameless atomic absorption spectrometry (AMA 254, SYMALAB, France). Detection limit of the instrument is 1.4 ng g_{dw}^{-1}. The validity of the analytical method is checked against three biological reference materials (NRCC-CNRC, Ottawa, Canada): TORT-2 (lobster hepatopancreas), DORM-2 (dogfish muscle), and DOLT-2 (dogfish liver). Hg values are consistently within the published certified ranges. Measurements, expressed in dry weight (dw) by the machine, are converted into wet weight (ww) following the relationship determined by Maury-Brachet et al. (2006). All the Hg concentration values presented in this manuscript are fish muscle concentration values, and they are expressed in ww.

Statistical analysis

Descriptive values presented in this study correspond to the mean value of a population and its associated standard error. Student's *t* test is used to determine the difference between mean mercury concentrations as well as mean standard lengths. If not specified, the significance is calculated at the 95% confidence level. To estimate the risk probability ("Inter-river comparison" section), the kernel density estimation method (Parzen 1962) is used.

Levels of contamination

Three levels of mercury contamination in fish muscle are determined based on the WHOsl: (1) lower than the safety limit of 0.5 mg kg^{-1}, (2) between 0.5 and 1 mg kg^{-1}, and (3) higher than 1 mg kg^{-1} which correspond to at least twice the safety limit value. For each of the 51 sectors, the percentage of the three levels of mercury contamination is determined and presented as pie charts. To facilitate the description of spatial distribution of mercury contamination, sectors are regrouped into different geographical clusters of similar levels of contamination and/or same contamination pressure origins.

Results and discussion

Inter-river comparisons

Mean standard lengths of fish sampled at each sampling sectors (mean values ± SE) and corresponding mean mercury concentration (mean values ± SE) are presented in Table 1. Visual representations of these values via box plots are presented in Fig. 3. The number of fishes per river is highly variable and depends on (i) the river length, (ii) the type of program that was in charge of sampling it, and (iii) the anglers' willingness.

Fish's size has an influence on the level of contamination. Box plots on fish's standard length (Fig. 3a) indicate that fish length distributions of the six rivers are not significantly different. The homogeneous fish length distribution between the six rivers leads to assume that mercury concentration variations are mainly related to the mercury contamination in the environment. The six rivers can be divided into three clusters (three different colors on Fig. 3b). Mean values of Maroni, Mana, Comté, and Approuague fish Hg concentrations are not statistically different at the 95% confidence level. These mean values are statistically different from the Oyapock river value, except for Comté. Fishes from the Sinnamary river present a mean Hg value significantly higher than all of the other ones. Statistics using median values are similar than for mean values. There is no river with mean fish Hg concentration values below the World Health Organization (WHO) safety limit of 0.5 mg kg^{-1} (Fig. 3b). For the remaining of the manuscript, WHO safety limit is referenced as WHOsl.

Probabilities of catching a fish below the WHOsl are presented in Fig. 4. Probability values are heterogeneous, going from 46% for the Oyapock river to 96% for the Sinnamary river. There is a contamination gradient between the rivers, with Oyapock being the less contaminated followed by Comté, Maroni, Approuague, Mana, and Sinnamary. Probabilities presented in Fig. 4 highlight a serious health risk for populations that currently catch fish in these rivers. Almost all the fishes from the Sinnamary river present Hg concentrations higher than the WHOsl. Even in the Oyapock river that has the lowest mean mercury concentration, one has nearly 50% risk of catching a fish with Hg concentration higher than the WHOsl.

Spatial distribution of mercury concentrations in fishes from each river

Table 2 presents the number of individuals, their standard lengths, and corresponding mean mercury concentrations per sectors. For the remaining of this manuscript, every occurrence of mercury concentrations refers to the mean fish muscle mercury concentration per sector. Fish mercury contaminations are regrouped into three different color-coded mercury

Table 1 Average Hg concentrations (mg kg^{-1} ww) and average standard length of fishes for each river; value ± standard error (SE); *Number* number of samples, min and max

River	Number	Mercury concentration (mg kg^{-1} ww)				Standard length (cm)			
		Mean	SE	Min	Max	Mean	SE	Min	Max
Maroni	154	0.671	0.026	0.181	2.221	54.86	0.78	40.0	80.0
Mana	64	0.675	0.030	0.312	1.326	59.11	1.11	41.0	76.0
Sinnamary	168	1.025	0.028	0.330	2.180	58.90	0.76	40.5	89.0
Comte	78	0.624	0.036	0.239	1.833	60.19	1.37	40.0	89.0
Approuague	57	0.684	0.044	0.118	1.690	59.21	1.44	40.0	80.0
Oyapock	54	0.548	0.036	0.215	1.657	z54.83	1.44	40.0	85.0
Total	575	0.758	0.015	0.118	2.221	57.66	0.43	40.0	89.0

concentration levels, based on the WHOsl: (1) lower than 0.5 mg kg^{-1} (green level), (2) between 0.5 and 1 mg kg^{-1} (red level), and (3) higher than 1 mg kg^{-1} (black level). For each sector, the percentage of the three mercury concentration levels are determined and presented in Fig. 5. This figure presents a visual estimation of risk levels that the population is exposed to when eating piscivorous fishes. Geographical clusters of sectors presenting similar level of contamination distributions and similar contamination pressure are described along each river.

Maroni river

The Maroni river, the westward river of French Guiana, is the longest of the six rivers (Barret and Vendé 2002). This river has 15 sectors (1–15). There are a large number of both former and current gold mining sites along the Maroni river (Carmouze et al. 2001). Fish mercury concentrations in this river present a wide range of values going from 0.3 to 0.88 mg kg^{-1} (Table 2). The upstream Maroni sector, "Source Litani" (1), has more than three fourths of its fishes with the red level, the rest in green level, and no fish was found with the black level of mercury concentration. The mean fish Hg value of this sector is close to the WHOsl (0.54 mg kg^{-1}). This sector can be defined as pristine sector for mercury contamination as there are no gold mining activities neither urban areas (Guedron et al. 2009). Sectors "Happapota" (2), "Tampok" (3), "Waki × Tampok" (4), "Antecume Pata" (5), "Wanapi" (6), and "Limonade × Couleuvre" (7) are regrouped into the upstream Maroni river cluster. This cluster presents high mercury concentration values ranging from 0.59 to 0.73 mg kg^{-1}, probably due to current gold mining activities (gray hatched areas on Fig. 5). A high increase in illegal gold mining activities is currently observed in this area (PAG 2017). Additionally, there is a large gold mining activity near the border with Surinam that contaminates the upstream Maroni sector via two main tributaries: "Oulemani" and "Lowé" (Laperche et al. 2008; Dezécache et al. 2017; Rahm et al. 2017). Sectors "Grand Inini" (8), "Eau Claire × Dupouy" (9), "Petit Inini" (10), "Papaïchton" (11), "Abounami" (12), "Mouchounga" (13), and "Sparouine" (14) are part of a cluster presenting very high mean Hg values, ranging from 0.7 to 0.9 mg kg^{-1}. This cluster is located in an intense gold mining area. The last Maroni sector, "Saint Laurent du Maroni" (15), is the only sector presenting solely green level of contamination. This sector is the downstream sector, and the water going up from the estuary might induce a dilution, which decreases the contamination level (Laperche et al. 2014).

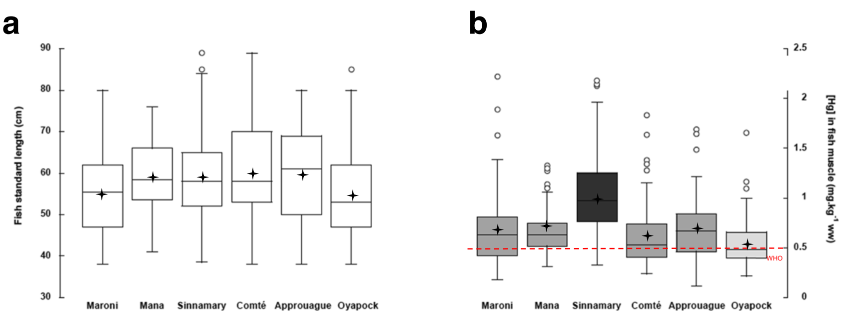

Fig. 3 Box plots of **a** *Hoplias aimara* standard length (in cm) and **b** mercury concentration in *Hoplias aïmara* muscles (in mg kg^{-1} ww) from six Guiana rivers. The red dotted line indicates the safety limit value established by the World Health Organization (0.5 mg kg^{-1} ww). Clusters with the same color code are not significantly different at the 95% confidence level. Black lines within the boxes mark the median, and stars represent the mean

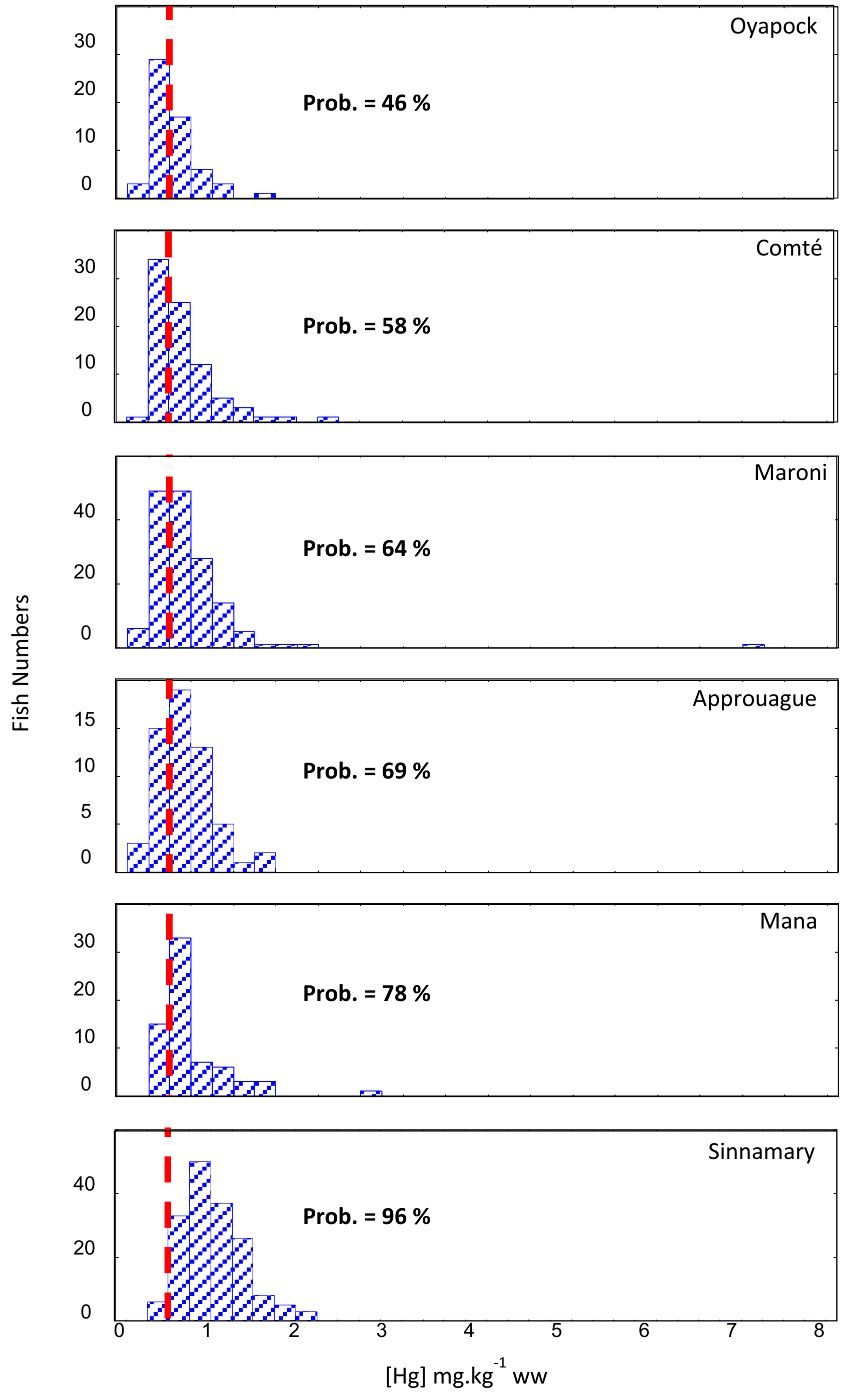

Fig. 4 Distribution of mercury concentrations ([Hg] in mg kg^{-1} ww) measured in fish muscles from six French Guiana rivers in mg kg^{-1}. The dashed red line represents the World Health Organization (WHO) guideline for mercury concentration (0.5 mg kg^{-1} of wet weight). Prob. values represent probabilities of fishing a *Hoplias aimara* with a mercury concentration higher than the WHO recommendation

Mana river

The Mana river has six sectors (16–21), with mercury concentrations ranging from 0.52 to 0.8 mg kg^{-1}. Sector "Repentir" (16), the most upstream Mana's sector, has a high Hg concentration of 0.71 mg kg^{-1}. This region is impacted by illegal gold mining activities (PAG 2017). The cluster regrouping sectors "Bois Courroné" (17), "Arouani" (18), and "Deux Fromagers" (19) is also impacted by current gold mining activities. Sector "Kokioko" (20) presents a low mercury concentration (0.57 mg kg^{-1}), and only few gold mining sites are present in this sector (Laperche et al. 2007). Sector "Angoulème" (21), the most downstream Mana's sector, has a mercury value (0.52 mg Hg kg^{-1}) close to the WHOsl. Similarly as sector 15 ("St Laurent du Maroni"), this low Hg value might be due to a dilution process operating near the estuary area.

Table 2 Average Hg concentrations (mg kg^{-1} ww) and average standard length of fish for each site; value ± standard error (SE); *Number* number of individuals

River	Sector no.	Name of the sector	Number	Mercury concentration (mg kg^{-1} ww)		Standard length (cm)	
				Mean	SE	Mean	SE
Maroni	1	Source Litani	10	0.54	0.06	59.80	3.97
	2	Happapota	11	0.71	0.16	52.15	3.34
	3	Tampok	10	0.73	0.09	53.70	2.51
	4	Waki × Tampok	15	0.59	0.08	54.60	2.08
	5	Antecume Pata	10	0.67	0.07	50.80	1.76
	6	Wanapi	5	0.85	0.18	60.70	2.62
	7	Limonacle × Couleuvre	12	0.58	0.05	57.33	2.49
	8	Grand mini	23	0.69	0.06	59.35	1.76
	9	Eau Claire × Dupouy	11	0.88	0.13	53.68	2.63
	10	Petit mini	6	0.81	0.12	57.00	3.41
	11	Papaichton	12	0.74	0.10	49.83	2.89
	12	Abounami	10	0.74	0.09	58.10	4.51
	13	Mouchounga	5	0.71	0.08	48.80	4.40
	14	Sparouine	6	0.68	0.06	56.00	3.28
	15	St Laurent du Maroni	8	0.30	0.03	47.00	2.31
Mana	16	Repentir	13	0.71	0.07	60.96	2.58
	17	Bois Courrone	12	0.78	0.07	61.25	2.68
	18	Arouani	12	0.67	0.06	63.54	1.94
	19	2 Fromagers	8	0.80	0.09	59.50	2.10
	20	Kokioko	8	0.57	0.05	58.00	3.16
	21	Angoulenne	11	0.52	0.06	50.27	2.05
Sinnamary	22	Deux Roros	9	0.91	0.10	59.39	3.85
	23	Saut Takari Tante	20	0.95	0.06	61.33	1.14
	24	Vata	11	0.79	0.08	60.53	3.35
	25	Leblond	46	1.00	0.08	58.23	2.24
	26	Koursibo	22	0.94	0.05	56.02	1.43
	27	Saut Lucifer	13	1.11	0.07	60.69	0.97
	28	Saint Eugene	19	1.20	0.11	55.74	1.85
	29	Genipa	11	1.20	0.13	57.27	0.85
	30	Kerenroch	17	1.21	0.07	66.59	3.67
Comte	31	Belizon	9	0.56	0.09	69.00	5.02
	32	Galibi	12	0.52	0.05	60.50	2.57
	33	Lysis	7	0.55	0.07	60.71	3.70
	34	Bagot	27	0.50	0.03	51.04	1.32
	35	Cacao	12	0.68	0.11	60.25	2.51
	36	Pri-pri	11	1.08	0.12	74.73	2.39
Approuague	37	Sapokal	7	0.75	0.11	58.43	4.88
	38	Saut Japigny	9	0.94	0.09	60.78	3.19
	39	Saut Grand Kanori	4	0.63	0.24	62.25	5.31
	40	Couy	8	0.88	0.18	63.63	4.13
	41	Saut Grand Machikou	5	0.64	0.15	54.80	1.50
	42	Aratal × Japigny	15	0.55	0.04	55.00	2.64
	43	Dubai	4	0.74	0.10	65.25	2.39
	44	Regina	5	0.34	0.11	60.20	8.13
Oyapock	45	Trois Sauts	12	0.40	0.04	57.08	3.71
	46	Moulou Moulou	6	0.53	0.08	55.83	4.69
	47	Yanioue	7	0.75	0.09	54.86	4.22
	48	Saut Camopi	8	0.65	0.09	53.25	2.22
	49	Sikini	6	0.76	0.19	63.83	2.06
	50	Saut Maripa	5	0.61	0.15	52.60	5.95
	51	Gabaret	12	0.45	0.05	50.67	3.06

Sinnamary river

The Sinnamary river has nine sectors (22–30) that all have very high Hg concentration values ranging from 0.8 to 1.2 mg kg^{-1}. The Hg concentration in 96.4% of fishes is above the WHOsl. The Sinnamary river sectors are regrouped into five clusters: (A) the upstream east: "Deux Roros" (22) and "Saut Takari Tanté" (23), (B) the upstream west: "Leblond" (25) and "Koursibo" (26), (C) the Petit Saut Lake east: "Vata" (24), (D) the Petit Saut Lake west: "Saut Lucifer" (27), "Saint Eugène" (28), and "Génipa" (29), and (E) the downstream: "Kérenroch" (30). "Takari Tanté" (23) and "Lucifer falls"

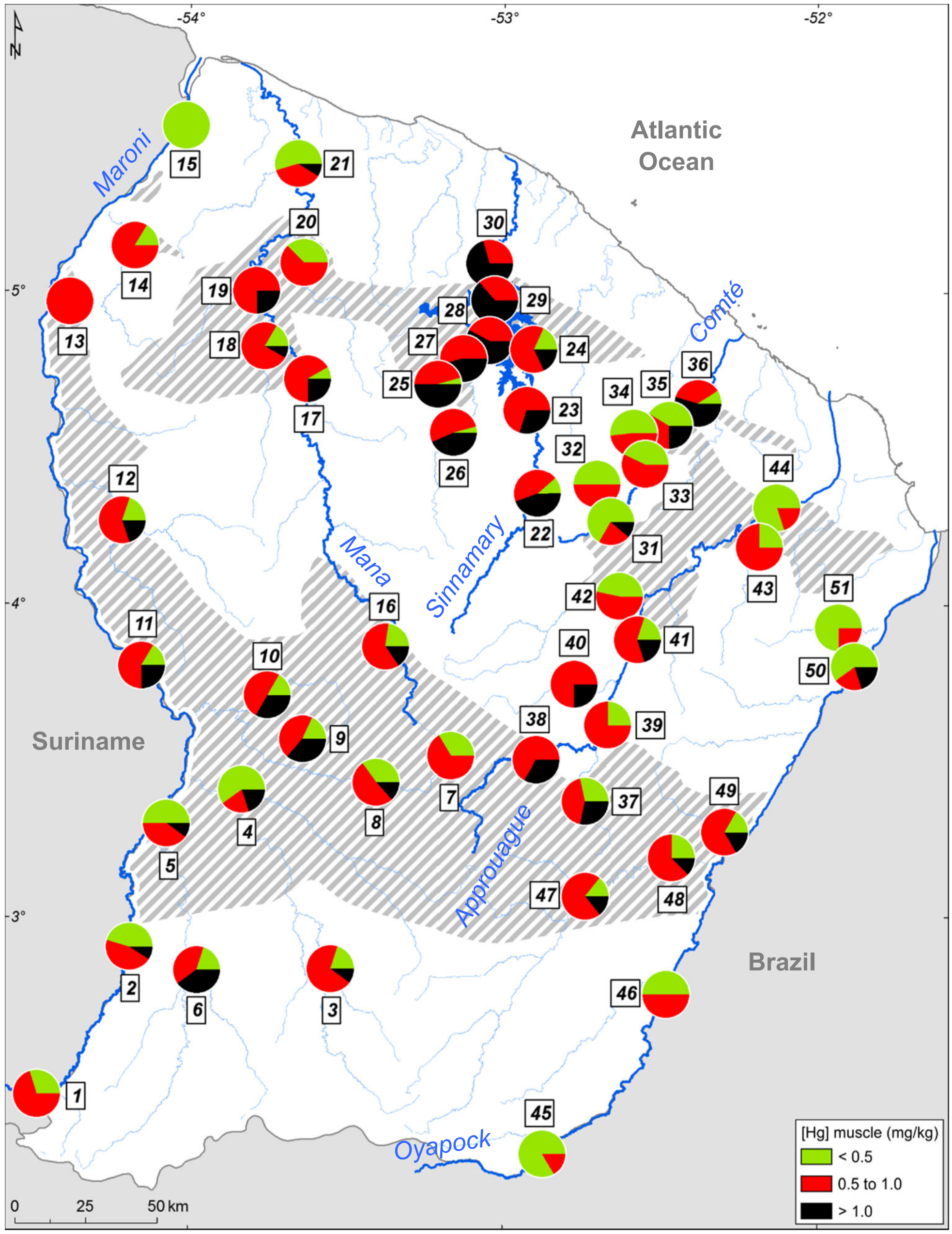

Fig. 5 Map of the level of mercury contamination along the six main Guiana rivers. For each sector, the proportion of the three levels of contamination (green < 0.5 mg kg^{-1}; red 0.5 to 1.0 mg kg^{-1}; black > 1.0 mg kg^{-1}) were calculated and reported as pie charts. Gray hatched areas correspond to areas under current gold mining activities

(27) separate the upstream east and upstream west clusters from the Petit Saut Lake. A hydroelectric dam separates Petit Saut Lake from the downstream region.

In the lake region, clusters C and D present high mean mercury contaminations. During the establishment of the hydroelectric dam in 1995, 360 km^2 of forest was flooded, which induced an anoxia of the water column (up to 30-m depth). The anoxia lead to an important production of MeHg by the biofilm that settled on various underwater substrates such as tree trunks, leafs, and sediments (Charlet and Boudou 2002; Coquery et al. 2003; Dominique et al. 2007; Muresan et al. 2008; Huguet et al. 2009; Castilhos et al. 2015). The high MeHg production explains the high Hg concentrations found in *H. aimara* from the lake region (clusters C and D).

The western upstream region, cluster B, presents very high Hg concentrations (superior to 1 mg kg^{-1}). This cluster receives water coming from the vast, currently operating, gold mining site of Saint Elie (99 km^2; SMSE 2016) that is surrounded by illegal gold mining activities.

The eastern upstream region, cluster A, presents higher Hg concentrations than expected since this area is not under current gold mining activities. A long-term monitoring of fish mercury concentrations at “Deux Roros” (1992–2004) indicates that fish mercury concentrations increase drastically in

2000, and stay high since then (Dominique 2006). This study concluded that the decamillennial flooding event of 2000 is responsible for this upstream contamination. Their hypothesis is that this flooding might have broken natural barriers (sectors 23 and 27) separating upstream rivers from the lake. This hypothesis explains the similar Hg concentrations observed between clusters A and C.

The downstream region, cluster (E), presents higher Hg concentrations (1.2 mg kg^{-1}) than expected for a downstream sector. This contamination has the same origin than the upstream contamination observed in cluster A. During the 2000 flooding, a large quantity of water, and therefore of biofilm, was discharged downstream leading to the fish contaminations of the downstream Sinnamary river (Dominique 2006). In 2006, the MeHg production in the downstream area was as important as in the lake area (Muresan Paslaru 2006; Muresan et al. 2008). This downstream MeHg production is responsible for the high concentration (1.21 mg kg^{-1}) found at the sector E, "Kérenroch" (30).

Comté river

The Comté river has six sectors (31–36). Four sectors of relatively low Hg contaminations ranging from 0.5 to 0.56 mg kg^{-1}, "Bélizon" (31), "Galibi" (32), "Lysis" (33), and "Bagot" (34), were impacted by the first wave of gold mining activity (1850 to 1920), but no illegal gold mining activities have settled since (Laperche et al. 2014).

Sector "Cacao" (35) has a low mean Hg concentration of 0.68 mg kg^{-1} but presents black-level contaminations. This mean value is the result of intensive agriculture in this sector where soil leaching is very important (Grimaldi et al. 2008).

The downstream sector "Pri-pri" (36) has the highest Hg concentration value (1.08 mg kg^{-1}) of Comté river. This sector is located in a natural swamp area. The low depth and high anoxia conditions of the swamp are favorable to MeHg production leading to Hg bioaccumulation in piscivorous fishes (Benoit et al. 2001, 2003).

Approuague river

The Approuague river has eight sectors (37–44) of various contamination levels.

The upstream sectors "Sapokaï" (37), "Saut Japigny" (38), "Saut Grand Kanori" (39), "Couy" (40), and "Saut Grand Machikou" (41) present high mercury contamination levels. The upstream Approuague river area is the area that had, and still has, the more active illegal gold mining sites in Guiana (Laperche et al. 2014).

Sector "Arataï × Japigny" (42) presents a low mercury contamination of 0.55 mg kg^{-1}. This sector, like sectors 31 to 34 on the Comté river, is located on a former gold mining sector (Laperche et al. 2014).

Sector "Dubol" (43) has a high mean Hg concentration value (0.74 mg kg^{-1}); this contamination stems from former gold mining activities from a nearby area. As observed for sectors 15 and 21 on the Maroni and Mana rivers, respectively, the sector "Régina" (44) is under the tide influence. Mean fish muscle Hg concentration is very low (0.34 mg kg^{-1}) and below the WHOsl.

Oyapock river

The Oyapock river has seven sectors (45–51). The upstream sector, "Trois Sauts" (45), is located in an area without any gold mining activities and has a low mean mercury concentration value (0.40 mg kg^{-1}) below the WHOsl.

Sector "Moulou Moulou" (46) has a mean mercury concentration value slightly above the WHOsl (0.53 mg kg^{-1}), but no black-level contamination is observed. This sector is located in area without gold mining activities.

The cluster regrouping "Yanioué (début)" (47), "Saut Camopi" (48), and "Sikini" (49) has high Hg concentration values (0.61 to 0.76 mg kg^{-1}). This cluster is situated in an area impacted by gold mining sites since the start of the Guiana gold mining activities.

The sector "Saut Maripa" (50) has a mean Hg concentration value lower than the previously described cluster but still higher than the WHOsl (0.61 mg Hg kg^{-1}) with some black-level contaminations. From 1950 to 1980, this sector was under gold mining activities; however, gold mining activities stopped totally in 1980.

The sector "Gabaret" (51) has similar Hg contamination as the upstream sector (45). Sector 51 is located in a non-gold mining area and is under the tide influence.

Factors responsible for the spatial distribution of mercury contamination in French Guiana

There is a spatial distribution of mercury contamination in French Guiana with lower contaminations both upstream and downstream that surround an area of high mercury contamination (Fig. 5). This zonal distribution is influenced by both anthropogenic and natural factors.

Upstream pristine sectors

Only three upstream sectors (1, 45, and 46) present low mercury concentration values close to the WHOsl (~0.54, 0.40, and 0.53 mg kg^{-1}, respectively). The mean mercury concentration value and corresponding standard error in fishes for this area is 0.471 ± 0.032 mg kg^{-1}. These sectors were defined as pristine sites in a sediment contamination study (Laperche et al. 2014).

Contaminated sectors

Anoxia and gold mining activities are the two main factors responsible for the high mercury concentration in fish muscles.

Anoxia

Anoxia is the first factor responsible for Hg contamination in Guiana river fishes. Sectors under anoxia conditions present the highest Hg contaminations in French Guiana, with a mean value and standard error of 1.029 ± 0.027 mg kg^{-1}, which corresponds to 2.18 times the mean Hg concentration of pristine areas. In anoxia areas, the lack of oxygen promotes the development of sulfate-reducing bacteria responsible for the mercury methylation (King et al. 2000; Acha et al. 2005; Winch et al. 2009) which leads to the production of methylmercury (Benoit et al. 2001, 2003; Castilhos et al. 2015; Marrugo-Negrete et al. 2015). Anoxia areas are found in natural swamp areas as well as in lakes resulting from the creation of hydroelectric dams. In French Guiana, natural swamp area (sector 36) presents a high mercury concentration value, more than twice the value of the pristine areas. The hydroelectric dam located on the Sinnamary is responsible of the high mercury contamination of all the sectors surrounding the lake (22–30). The lake construction created an anoxic area leading to high MeHg productions. Establishment of hydroelectric dams has already been found responsible of similar mercury contaminations (Friedl and Wüest 2002; Boudou et al. 2005; Kasper et al. 2014; Castilhos et al. 2015). Sectors under the influence of the hydroelectrical dam are two to three times more contaminated than the pristine areas.

Gold mining activities

Illegal gold mining activities are the second most important factor controlling Hg contamination in Guiana river fishes and the one with the most damaging consequences. The mean Hg concentration value and corresponding standard error of gold mining areas is 0.717 ± 0.020 mg kg^{-1}, which corresponds to 1.52 times more the mean Hg concentration value of the pristine sectors.

Due to high gold mining activities along the Mana, Sinnamary, Comté, and Approuague rivers, no pristine sectors are identified on them. Sectors located in current gold mining areas present Hg concentration values from 1.5 to 2 times more than the pristine area value. Contaminations induced by illegal gold mining activities present therefore lower values than the one induced by the hydroelectric dam but has a higher impact on human populations and especially on native populations. In French Guiana, there are two areas where native populations are living in self-subsistence (hunting, fishing, and farming): one on the Maroni river (sector 5), a sector currently under illegal gold mining activities, and one on the Oyapock river (sector 45), a sector defined as pristine for mercury contamination. Analyses of mercury concentrations in native population hairs indicate that the population living on the sector 5 presents mercury contamination twice as much as the population living on the sector 45 (Cardoso et al. 2010; BAZAG 2007; Pignoux et al. submitted).

Illegal gold mining activities are without a doubt a dangerous source of contamination. On a positive note, former gold mining sectors are 1.30 times less contaminated than sectors under current illegal gold mining activities (0.550 and 0.717 mg kg^{-1}, respectively). This gives some hope that if illegal gold mining activities stop, sites might recover from those contaminations.

Downstream sectors under tide influences

Downstream areas (sectors 15, 21, 44, 51) present mercury concentration values lower than sectors located upstream of them, and their values are close to the pristine area values. The tide plays a tempering role on the mercury contamination since a natural dilution is happening in estuary areas. The mean Hg concentration and corresponding standard error of the estuarine sectors is 0.424 ± 0.032 mg kg^{-1}, value similar to the mean Hg concentration of the pristine sectors.

Temporal evolution of mercury contamination

Two sectors upstream of the Oyapock river, "Saut Camopi" (48) and "Trois Sauts" (45), are compared to estimate the temporal evolution of Hg contamination between 2005 (data presented in "Spatial distribution of mercury concentrations in fishes from each river" section) and 2014 (Fig. 6). "Saut Camopi" (48) is located in a gold mining area, and "Trois Sauts" (45) is located in a pristine area without any known gold mining activities. There are no significant differences in the standard length of fishes sampled in the two sectors nor between fishes sampled in 2005 and 2014 (Fig. 6a). In 2005, the pristine and the gold mining areas' mean Hg concentration values are 0.40 mg kg^{-1} ww and 0.72 mg kg^{-1} ww, respectively. In 2014, the pristine and the gold mining areas' mean Hg concentration values are 0.42 mg kg^{-1} ww and 0.65 mg kg^{-1} ww, respectively. There are significant differences in Hg mean concentrations between those two sectors (Fig. 6b). There are, however, no significant differences in Hg concentrations between 2005 and 2014 for neither the pristine nor the gold mining area. The environment might need more than 10 years to clean itself from the contamination.

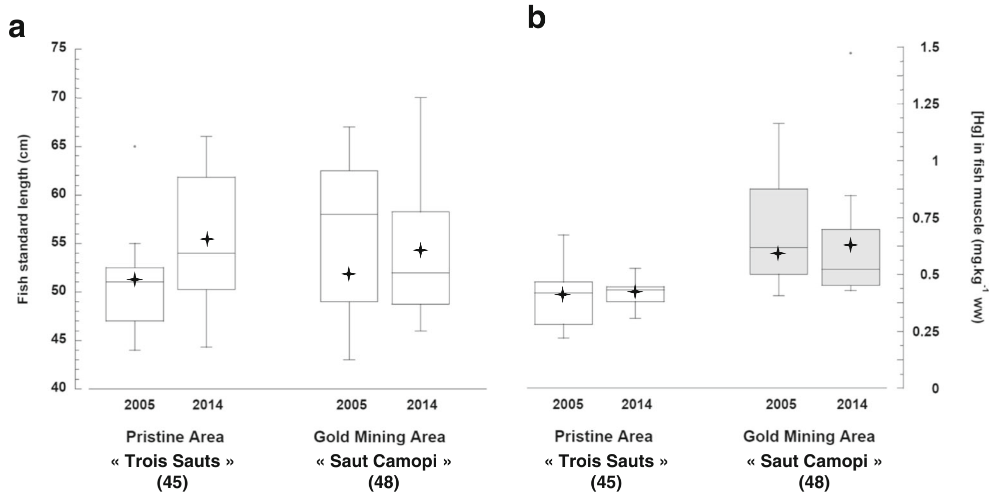

Fig. 6 Box plots of **a** *Hoplias aimara* standard length (in cm) and **b** mercury concentration in *Hoplias aimara* muscles (in mg kg^{-1} ww) in two different areas (gold mining area—"Saut Camopi" (48) and pristine area—"Trois Sauts" (46)) for two different years (2005 and 2014). Clusters with the same color code are not significantly different at the 95% confidence level. Black lines within the boxes mark the median, and stars represent the mean

Conclusion

The study of Hg concentrations in fish muscles permits to assess the contamination level of the six main French Guiana rivers using the bioindicator fish *Hoplias aimara*. The Oyapock river presents the lowest Hg contamination with mean Hg concentration value below the WHOsl. Maroni, Mana, Comté, and Approuague rivers present Hg concentrations right above the WHOsl. The Sinnamary river presents the highest level of contamination with Hg concentration mean value more than twice the WHOsl value. Level of contaminations of individual sectors and/or clusters for each river is not homogeneous. A zonal spatial distribution of Hg contamination is observed in French Guiana. A contaminated area is surrounded by both upstream and downstream areas of lower Hg contaminations. This spatial distribution is influenced by both anthropogenic and natural factors. The low Hg contamination observed upstream corresponds to pristine areas without any influence of gold mining activities. The low Hg contamination observed downstream is a consequence of the natural dilution happening in estuary areas. Anoxia and gold mining activities are the two main factors responsible for high mercury concentrations in fish muscles. While levels of mercury contaminations are higher in anoxia areas (natural swamp and hydrological dam), contaminations induced by gold mining activities present the most harmful consequences to human populations. Native populations living near illegal gold mining sites present levels of contaminations twice higher than populations living in pristine area.

The two Oyapock sectors that were analyzed 10 years apart do not present any significant changes in Hg concentration. This highlights the fact that despite the ban of mercury for gold mining activities in French Guiana, the level of mercury did not decrease. The environment might need more than 10 years to recover from mercury contaminations. The spatial contamination map presented in this study might be useful for stakeholders to establish an environmental plan. Sectors where the native Amerindian population live should be monitored in order to prevent sanitary disasters.

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