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Abstract

In French Guiana, native population present high level of mercury contamination, which has been linked to the consumption of contaminated fishes. Gold mining activities in French Guiana are considered as the main source of contamination. The use of mercury in Guiana is prohibited since 2006, but unfortunately, illegal gold mining activities are still using it. The goal of this study is to undertake an inventory of mercury contamination levels of the six main Guiana Rivers and to established links between contaminated areas and gold mining activities. The selected species for this study is the ubiquitous piscivorous species *Hoplias aimara*. It is a perfect species for this study as it is (i) a good bio indicator of mercury contamination along freshwater food webs, (ii) easy to identify, (iii) representative of the fishing site as it makes only transverse migrations across short distances, and (iv) the most fished species by the native population for consumption. A total of 721 fishes from 134 discrete fishing sites were regrouped into 51 river sectors. Results from this study permits to rank the six main Guiana Rivers by their level of contamination with the Oyapock being the less contaminated river followed by Comté, Maroni, Approuague, Mana, and Sinnamary. The contamination is however not spatially homogenous along each river and 86 % of the sectors are contaminated and therefore present high risks for human health. A map of the different levels of mercury contamination is provided and areas on similar levels of contamination are highlighted. A temporal comparison of two sites a pristine area and a site under gold mining activities are compared ten years apart and unfortunately no decrease in mercury contamination is observed in the contaminated sector.

1. Introduction

Mercury (Hg) is naturally present in the environment through volcanism, soil erosion, and ocean degassing. However, human activities are the major actor in releasing Hg into the environment via power plants, incinerators, industrial activities, and goldmining (Streets et al. 2009; Driscoll et al. 2013; Kocman et al. 2017). Mercury is classified as 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°) which is presents under liquid and volatile forms, (ii) the oxidized divalent mercury (Hg2+) presents under free or complexed forms, (iii) the methylmercury ($CH3Hg^{+}$), 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 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 in river’s water could start as low as $ng.l^{-1}$ to $mg.kg^{-1}$ into piscivorous fishes’ muscles (Coquery et al. 2003; Maury et al. 2006; Castilhos et al. 2015; Bradley et al. 2017)

Goldmining activities are present all over the French Guiana territory. Those 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 1850, it has been estimated that more than 300 tons of Hg has been released into French Guiana gold-bearing soils (Picot et al. 1993; Bizi et al. 2005; Cottard et al. 2012; Laperche et al. 2014). Since 2006, the utilization of Hg is prohibited in French Guiana; therefore, legal goldmining sites stopped using it. Illegal goldmining activities, which represents more than 98 % (PAG 2017) of the total goldmining activities in French Guiana, continues to release Hg in the environment. This illegal goldmining activity, more than having disastrous impact on the environment is also directly harmful for the native population. A high exposure to Hg leads to serious health issues (Grandjean et al. 1994; Mergler et al. 2001, 2007). In French Guiana, studies have showed that native population present high level of Hg contamination (Cordier et al. 1997; Fréry et al. 2001; Cardoso et al. 2010; Fujimura M. 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 (Frery et al. 1999). The level of population contamination varies depending of the living area, which imply a spatial heterogeneity of fish Hg contaminations. Illegal goldmining activities have also strong impacts on the fishes’ health, biomass and diversity: (i) fishes present Hg contamination, (ii) increase water turbidity (Laperche et al. 2014) which create hostile living habitat for fishes and therefore decrease their biomass and biodiversity (Brosse et al. 2011; Costa et al. 2012; Tudesque et al., 2012); (iii) deforestation due to gold mining is 4300 hectares in French Guiana (Dezécache et al. 2017) (iv) illegal gold miners are settling near the rivers and therefore increasing the fishing pression (Castilhos et al. 2015; Thomassin et al. 2017). For the remaining of the manuscript, references to goldmining activities mean, indeed, illegal ones.

It is of societal and environmental importance to determine the level of Hg contamination in fish in the French Guiana territory. This will be helpful to the French Guiana community as well as environmental managers to pinpoint contaminated vs pristine fishing areas. In this context, we chose a bioindicator fish - Hoplias aïmara- to describe Hg concentrations from various sites along the six main French Guiana Rivers. The objectives of this study were: (1) to obtain a map of the Hg contamination repartition in this fish species in the whole territory, (2) to perform an inter-river Hg level comparison and possible causes of the contamination, (3) to evaluate the temporal Hg contamination evolution at two times period (2005 and 2014) by comparing two distinct sites (a pristine site and a goldmining-impacted site).

2. Materiel and methods

2.1. The chosen 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 on the six main watersheds in French Guiana. This species has a sedentary life; it only does traversal displacements and during the wet season, goes back to nearby creeks to catch prey (Planquette et al. 1996). This species is therefore mainly representative of the capturing site. It is also one of the most fished species by the native population for consumption, which will allow estimating the human risk stem from consuming those fishes. All these characteristics makes this species an excellent bioindicator of mercury contamination.

2.2. Sampling sites

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. A total of 134 discrete fishing sites along those six rivers were selected. Those sites were then regrouped into 51 sectors in order to (i) obtain enough individuals per location, (ii) average sites of similar fish-living conditions (territory of about 10km), and (iii) recover sites that are under similar level of contamination.

Fishing campaigns came from two distinct programs ran 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 the local population. More than one hundred sampling kits were hand out to volunteers (schools, free clinics, police stations, fisherman, and local researchers) with the necessary 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, 2 markers. An explanatory document described how to recognize the wanted fish species, and how to dissect fish muscles. On a provided index card, the volunteer 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 the volunteer work, French scientists organized four fishing campaigns during which, they recorded similar information on their samples that was 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 et Nijssen 1989, le Bail *et al.* 2000, Planquette *et al.* 1996).

The second program realized twelve campaigns were both fishes and sediments were sampled. Those campaigns targeted non-habited areas.

The local population used fishing traps (Dundee Shop – Cayenne) made of a large self-shoeing hook at the end of a steel rope. 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 were collected with 67 % (480 fishes) from the CNRS program and 33 % (241 fishes) from the French Geological Survey program (Laperche *et* al. 2007; 2014).

In addition to the data obtain on this study, we received data from another French program (RIMNES project: Mercury Isotopes 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 sector “Trois Sauts” and “Saut Camopi”. Those data will allow to have an estimate of the temporal evolution of fish contamination.

2.3. Data processing:

On the 721 fishes collected, size ranged from 10 to 110 cm (Fig. 2). The selected species for this study, *Hoplias aimara*, looks morphologically like *Hoplias malabaricus*. Fish species determination was possible thanks to already published studies of Guiana fishes (Boujard *et al.* 1997, Keith *et al.* 2000, Kullander et Nijssen 1989, le Bail *et al.* 2000, Planquette *et al.* 1996). Those 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 level of 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). There is, to our knowledge, no study on age determination on neither *Hoplias aimara* nor *Hoplias Malabaricus*. To exclude all the *H. malabaricus* individuals, fishes with standard length above 40 cm were not included in this study. There is a logarithmic relationship between length and Hg concentration in piscivorous fishes (Durrieu 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 above 90 cm (5 % of the end tail distribution: Fig. 2) and use a range of length from 40 to 90 cm. The total number of fishes remaining is 575.

2.4. Mercury analyses

For each fish, an aliquot of muscle was sampled and dried in a 45°C oven for 48 hours. Total Hg concentrations in fish muscles were 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 was checked during each series of measurements against three biological reference materials (NRCC-CNRC, Ottawa, Canada): TORT-2 (lobster hepatopancreas), DORM-2 (dogfish muscle), and DOLT-2 (dogfish liver). Hg values were 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 expressed in ww.

2.5. Statistical testing

Descriptive values presented in this study correspond to the mean value of a population and its associated standard error. Student t-test is used to determine the difference between mean mercury concentrations as well as mean standard length. If not specified, the significance is calculated at the 95% confidence level. To estimate the risk probability (section 3.1.), the Kernel density estimation method (Parzen E. 1962) was used.

3. Results and discussion

3.1. Inter-Rivers comparisons

Mercury concentration and standard length values of *Hoplias aimara* were averaged by rivers (Table 1). A visual representation of those values *via* boxplots are presented in Fig. 3. The number of fishes per river is highly variable and dependent of (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. Fishes outside the 40-90 cm-standard-length were removed from the study (see section 2.3). Box plot on fish's standard length (Fig. 3.a) indicates that there is however, no significant difference between fish population lengths from the six different rivers. Mercury concentration variations are therefore only related to the mercury concentration in the environment. The six rivers can be separated into three clusters (three different colors on Fig. 3.b). Mean values of Maroni, Mana, Comté, and Approuague Hg concentration are not statistically different at the 95% confidence level. Mean mercury values from individuals of this cluster are statistically different from Oyapock River, except for Comté. Fishes from the Sinnamary River present the higher mercury concentration values, with a mean value significantly different form all the other five river mean mercury values. Statistics using median values are similar than for mean values. There is no River with mean fish mercury concentration values below the World Health Organization (WHO) safety limit of 0.5 mg.kg^{-1} (Fig. 3.b). For the remaining of the manuscript, WHO safety limit will be referenced as WHOsl.

Probabilities of catching fishes 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 highlights a serious health risk for population fishing in these rivers. Almost all the fishes from the Sinnamary River present mercury concentration higher than the WHOsl. Even for the Oyapock River, that has the lowest mercury concentration; one has nearly 50% chance to catch a fish with Hg concentration higher than the WHOsl.

3.2 Spatial distribution of mercury concentrations from each river

This section discusses the spatial distribution of Mercury concentrations along the six main French Guiana Rivers starting by the upstream sectors. Table 2 presents the number of individuals, their mean Mercury concentrations and standard lengths, per sectors. Fig. 5 displays the distribution per sectors of three different mercury concentration levels, based on the WHOsl: (1) lower than the safety limit of 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) which correspond to at least twice the safety limit value. Those groups permits a visual estimation of risk levels that the population is exposed to by eating piscivorous fishes. Clusters of similar level of contamination distributions are described along each river.

Maroni River

The Maroni River, the westward river of Guiana, is the longest of the six Rivers (Barret and Vendé 2002). This river has 15 sectors. There is a large number of both former and current gold mining sites along the Maroni River (Carmouze et al. 2001). Mercury concentrations in this river presents a wide range of values going from 0.3 to 0.88 $mg.kg^{-1}$ (Table2). The upstream Maroni sector, “Source Litani” (1), has more than ¾ of its fishes in the red level, the rest in green level, and no fishes are found with the black level of mercury concentration. The mean Hg value of this sector is closed to the WHOsl (0.54 $mg.kg^{-1}$). This sector can thus be selected as a reference-sector for mercury contamination as there is no gold mining activities neither urban areas in this sector (Guedron et al. 2009). Sectors “Litani” (2), “Tampock” (3), “Waki x Tampok” (4), “Antecume Pata” (5), “Wanapi” (6), and “Limonade x 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 gold mining activities. Illegal gold mining is currently observed in this area (PAG 2017), and there is many gold mining sites upstream of Antecum Pata. Those sites are located in Surinam, which contaminates the cluster *via* two main rivers: “Oulemani” and “Lowé” (Laperche et al. 2008; Dezécache C. et al. 2017; Rahm M. et al 2017). Sectors “Grand Inini” (8)”, “Eau Claire x Dupouy” (9), and “Petit Inini” (10) 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. Sectors “Papaïchton” (11), “Abounami” (12), “Mouchounga” (13), and “Sparouine” (14) is a cluster of high mercury values of about 0.7 $mg.kg^{-1}$ that might be a consequence of the intense upstream gold mining activities, as discussed right above. The last Maroni sector, “Saint Laurent du Maroni” (15), is the only sector that only presents green levels of contamination. This sector is the downstream sector, the water going up from the estuary might induce a dilution, which decrease the contamination level (Laperche et al. 2014).

Mana River

The Mana River has six sectors, with mercury concentration 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, ONF). The cluster regrouping sectors “Bois Courroné” (17), “Arouani” (18), and “Deux Fromagers” (19) is also impacted by high gold mining activities. Sector “Kokioko” (20), presents a low mercury concentration (0.57 $mg.kg^{-1}$), 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 $Hg.kg^{-1}$) closed 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.

Sinnamary River

The Sinnamary River has nine sectors, that all have very high Hg concentrations values ranging from 0.8 to 1.2 $mg.kg^{-1}$. 96.4% of fishes Hg concentrations are above the WHOsl. Those 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 west : “Vata” (24), (D) The Petit Saut Lake east: “Saut Lucifer” (27), “Saint Eugène” (28), and “Génipa” (29), and (E) The downstream: “Kérenroch” (30). Takari Tanté and Lucifer falls separate the upstream east and upstream west clusters from the Petit Saut Lake. A hydroelectric dam separates Petit Saut Lake from the downstream region.

The cluster A presents higher Hg concentration than expected since this area is not under current gold mining activities. Data from Dominique (2006) showed that during the decamillennial flooding event of 2000, there were a migration of fishes from the upstream to Lake Petit Saut and *vice versa*. This explains the similar Hg concentrations observed between clusters A and C, as well as between clusters C and D. Cluster B presents very high Hg concentrations (superior to 1 $mg.kg^{-1}$). This cluster receive water coming from the vast gold mining site of Saint Elie (99 km^2; SMSE 2016).

The downstream cluster (E), presents very high Hg concentration (1.2 $mg.kg^{-1}$), higher than what is expected for downstream sectors.

The join action of the 2000 flooding and the hydroelectric dam are responsible for the high Hg contamination of the Sinnamary River (Dominique 2006). During the establishment of the hydroelectric dam in 1995, 360 km^2 of forest were flooded, which induced an anoxia of the water column (up to 30 meter-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 et al. 2002; Coquery et al. 2003; Dominique et al. 2007; Muresan et al. 2008; Huguet et al. 2009; Casthilos et al. 2015). The high MeHg production explains the high Hg concentrations founds in *H. aimara* sampled in the Lake region (Dominique 2006). During the 2000 flooding, large quantity of water, and therefore of biofilm was discharged downstream. In 2006, the MeHg production in the downstream area was as important as in the Lake area (Muresan et al. 2008). This downstream MeHg production is responsible for the high concentration reaches (1.21 $mg.kg^{-1}$) found at the sector E,“Kérenroch” (30).

Comté River

The Comté River has six sectors of relatively low Hg contamination, except sector “pri-pri” (36). Sectors “Bélison” (31), “” Galibi” (32), “Lysis” (33), and “Bagot” (34), located on gold mining area, have lower Hg concentrations than gold mining sectors from both Maroni and Mana.

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

The downstream sector “Pri-pri” (36) has a higher Hg concentration value (1.08 $mg.kg^{-1}$). This sector is located in a swamp area of low depth and with high anoxia period, favorable to MeHg production and thus leading to Hg bioaccumulation in piscivorous fishes (Benoit et al. 2001, 2003).

Approuague River

The Approuague River has height sectors various levels of contamination.

Sectors “Sapokaï” (37), “Saut Petit Japigny” (38), “Saut grand Kanori” (39), “Couy” (40), and “Saut Grand Machikou” (41)” present the highest mercury contamination levels observed in Guiana rivers as a result of gold mining activities.

The upstream area of the Approuague River is the area that had and still has the more gold mining activities in Guiana (Laperche et al 2014).

Gold mining activities had stopped near sector “Arataï x Japigny” (42) (Laperche et al. 2014), and indeed this sector presents mean Hg concentration closed to the WHOsl value, and no black level contaminations are observed.

Sector “Dubol” (43) has a high mean Hg concentration value (0.74 $mg.kg^{-1}$) with black level contaminations. This contamination stem from former gold mining activities in the 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 Hg concentrations in both sediments (Picot et al. 1993) and fish muscles (0.34 $mg.kg^{-1}$) are very low and below the WHOsl.

Oyapock River

The Oyapock River has seven sectors. The more 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. This cluster is situated in an area with gold mining activities since the start of the Guiana gold mining.

The sector “Saut Maripa” (50) has a Hg concentration 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 x Oyapock” (51) has similar Hg contamination as the most upstream sector. Sector 51 is located in a non-gold mining area and as a downstream sector is influenced by the tide.

3.3. Spatial distribution of mercury concentrations in French Guiana

Area with mean Hg concentration below the WHOsl that can be considered as pristine area

Only sectors 1 and 45 located upstream of Maroni and Oyapock, respectively have mean mercury concentration values below the WHOsl. Those are the only two sectors that are not under gold mining activities. Those sites were also defined as pristine for sediment contaminations (Laperche et al. 2014).

Area with mean Hg concentration below the WHOsl that are located under tide influences

Downstream areas (sectors 15, 21, 44, 51) present mercury concentration values lower than sectors upstream of them, this is due to the natural dilution happening in estuary areas.

Area with mean Hg concentration above the WHOsl that are located in swamps

Sector “Pripri” (36), located in a swamp area, present high mercury concentration values. Swamp area are natural producer of Mercury, since they are area of high MeHg production (Benoit et al., 2001, 2003; Catilhos et al. 2015; Marrugo-Negrete et al. 2015).

Area with mean Hg concentration above the WHOsl that are impacted by a hydroelectric dam

The hydroelectric dam located on the Sinnamary is the cause of the high mercury contamination of all the sections of this river: the lake, upstream from the lake, and downstream from the lake (Boudou et al. 2005; Dominique et al. 2007).

The Lake construction created an anoxic area leading to high MeHg productions. Climatic event such as the 2000 flooding dispatched the contamination both upstream and downstream. This contamination is still noticeable nowadays. Similar contamination as already been linked to the establishment of hydroelectric dam (Friedl and Wüest 2002; Kasper D. et al 2014, Castilhos et al. 2015).

Area with mean Hg concentration above the WHOsl that are under gold mining activities

Gold mining activities are the main factor responsible for Hg contamination in Guiana Rivers. All the sectors/clusters located in former and/or current gold mining areas present Hg concentration values above the WHOsl. Due to high gold mining activities along the Mana, Sinnamary, Comté, and Approuague Rivers, no pristine sectors have been identified on them. This validates that gold mining activities is the major cause of mercury contamination in Guiana. It is not possible to do a direct comparison with previously published studies because used different fish species than the one used in this study (Catilhos et al. 2015; Basto et al. 2016), or similar species but with a very wide range of fish lengths (Mol et al. 2001).

Temporal evolution of mercury contamination

Two sectors upstream of the Oyapock River are compared to estimate the temporal evolution of Hg contamination between 2005 (data presented in section 3.2.) and 2014 (Fig. 6). The first sector is located in a gold mining area: "Camopi" (48); the second sector is located in a pristine area, meaning an area without any known gold mining activities: "Trois Sauts" (45). There is no significant difference in the standard length between the two sectors as well as between 2005 and 2014 (Fig. 6.a). There are significant differences in Hg mean concentrations between those two sectors (Fig. 6.b). 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. However, there are no significant differences in Hg concentrations between 2005 and 2014 for neither the pristine nor the gold mining area.

Conclusion

The study of fish muscle Hg concentrations 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 contamination of individual sectors and/or clusters for each river are not homogeneous. Seven sectors out of 51 are below the WHOsl, which imply that more than 86 % of the sectors are contaminated and therefore present high risks for human health. Those levels of contaminations are dependent on both natural (swamp areas) and human (gold mining activities and hydroelectric dam) forcings.

The two Oyapock sectors that were analyzed ten years apart do not present any significant changes in Hg concentration. This sadly highlight the fact that despite the ban of mercury for gold mining activities in French Guiana, the level of mercury did not decrease. The spatial contamination map presented in this study might be useful for stakeholders to establish an environmental plan. Sector where live Amerindian population, the most vulnerable population, should be monitored in order to prevent sanitary disasters.

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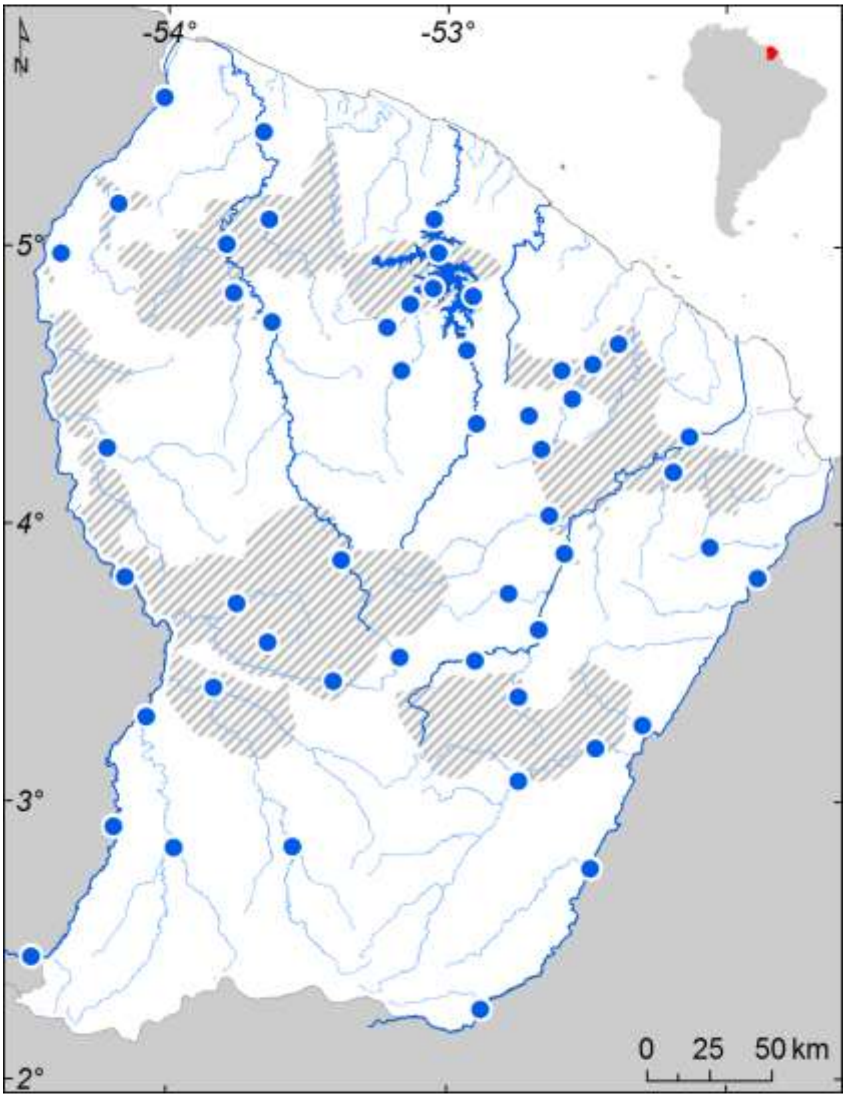

Fig. 1: location of selected sites on the 6 rivers of Guiana.

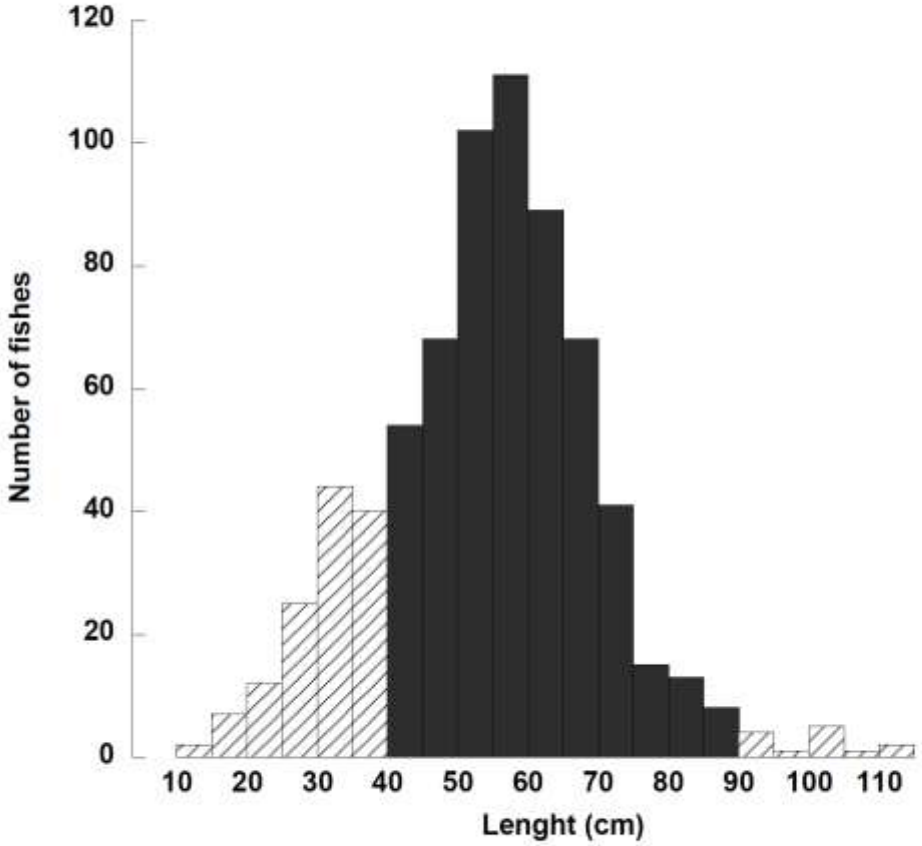

Fig. 2: Distribution of the length of sampled fishes. Fishes of size inferior to 40 cm were casted-off as they might include individuals from two different species. Fishes of size superior to 90 cm were removed, they can be considered as outliers (they represent the 2% of the end tail of the distribution).

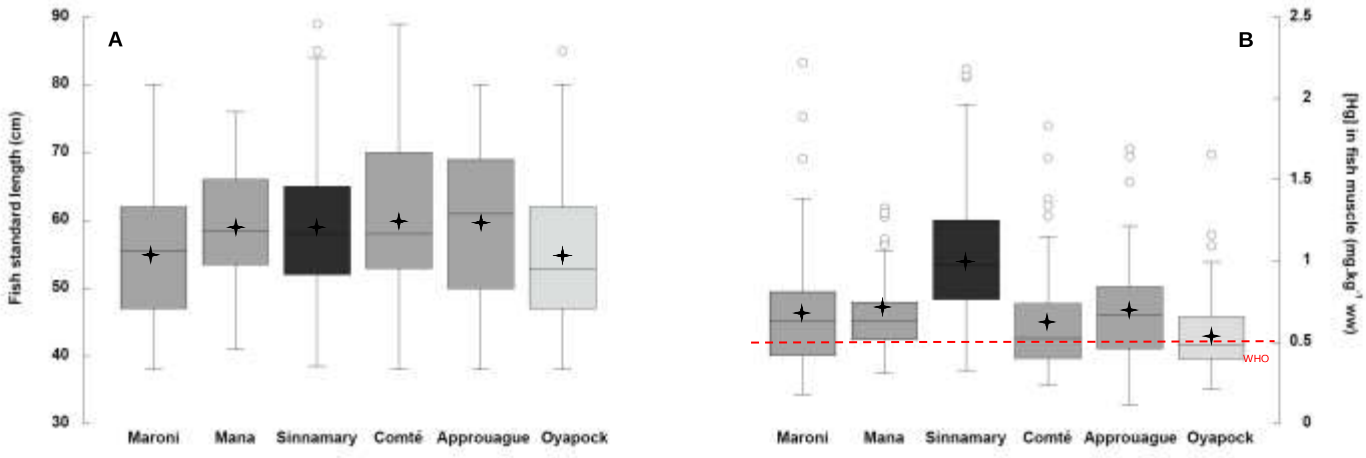

Fig. 3: Box Plots of (A) *Hoplias aimara* standard length (in cm) and (B) mercury concentration in *Hoplias aïmara* muscles (in mg.kg^{-1} ww) from six Guiana Rivers. The red dotted line indicates the safety limit value established by the World Health Organization (0.5 mg.kg^{-1} ww). Clusters with the 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.

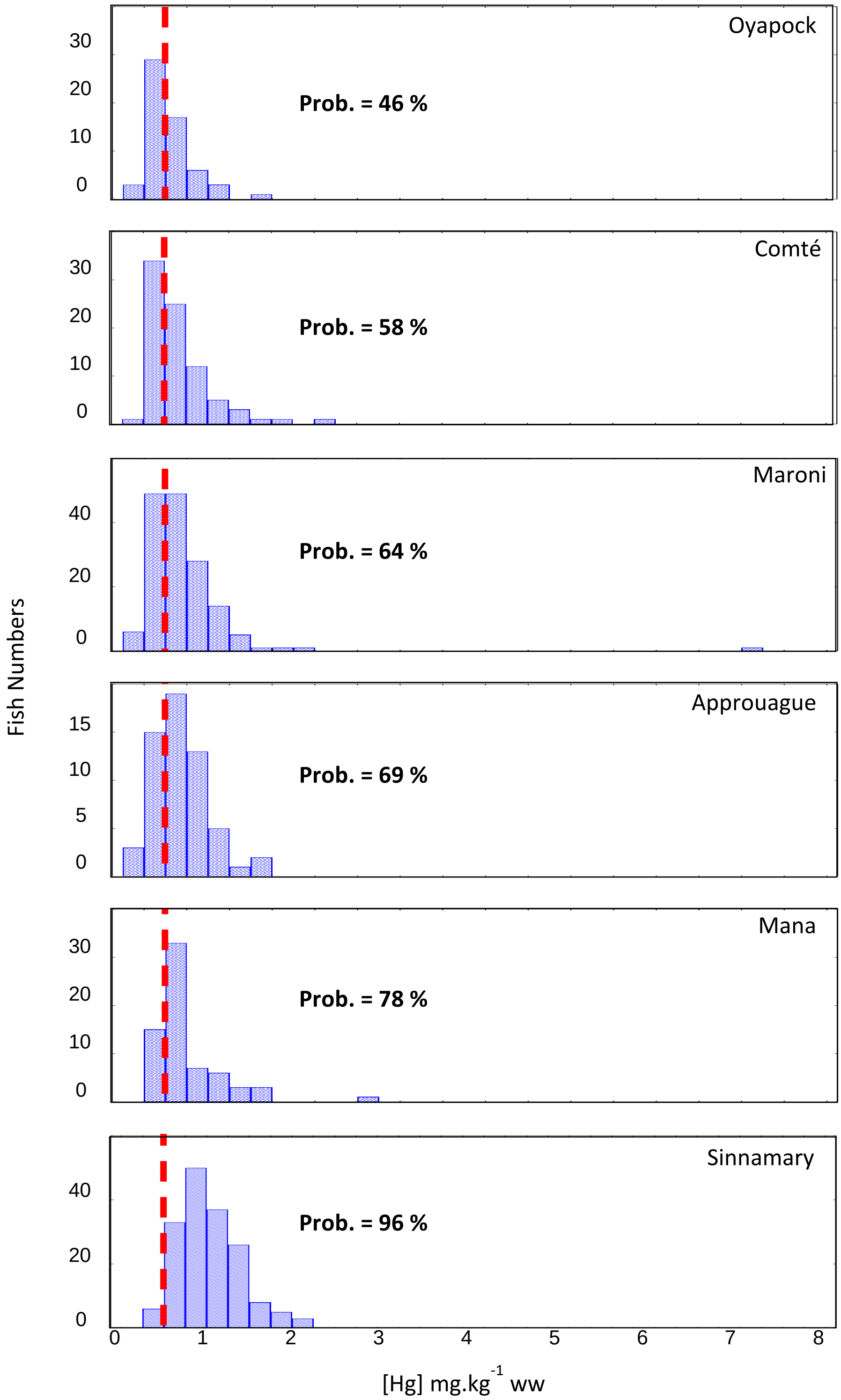

Fig. 4: Distribution of mercury concentrations ([Hg] in mg.kg^{-1} ww) measured in fish muscles from six French Guiana rivers in mg.kg^{-1}. The dashed red line represents the World Health Organization (WHO) guideline for mercury concentration (0.5 mg.kg^{-1} of wet weight). Prob. values represent probabilities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

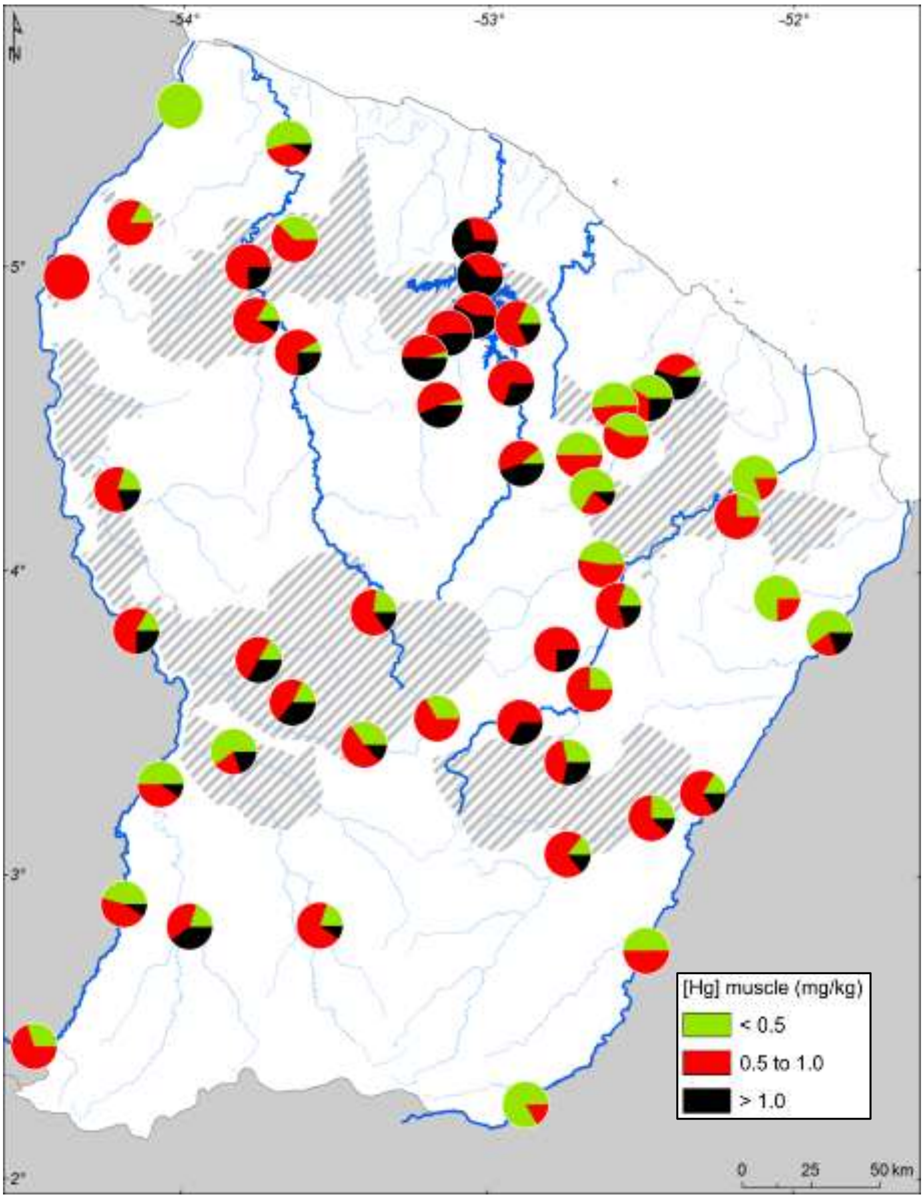

Fig. 5: Map of mercury concentration according to the 6 rivers

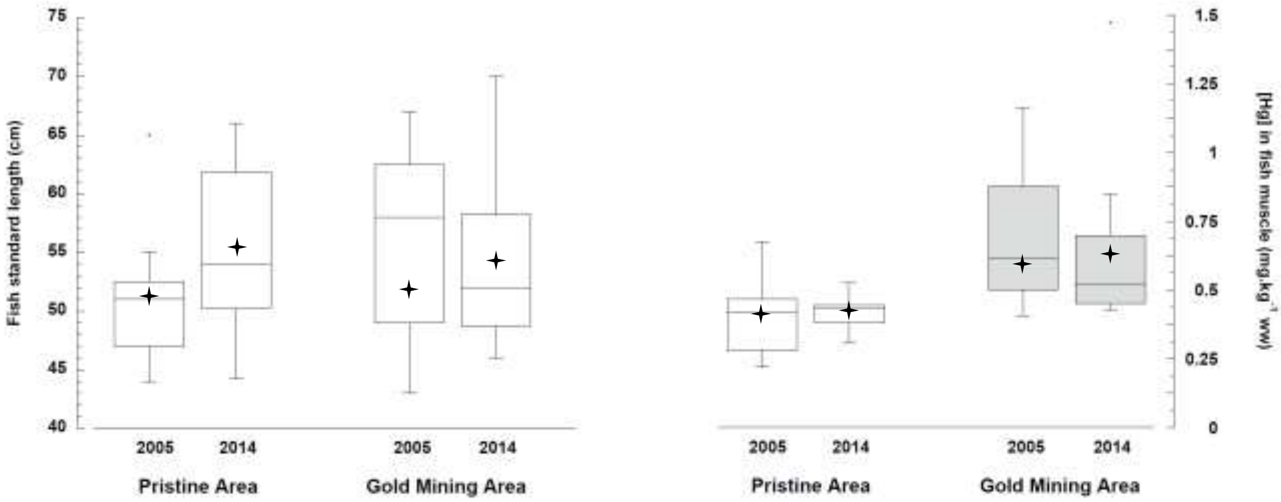

Fig. 6: Boxplots of (A) *Hoplias aimara* standard length (in cm) and (B) mercury concentration in *Hoplias aïmara* muscles (in $mg.kg^{-1}$ ww) two different areas (Gold Mining Area and Pristine Area) for two different years (2005 and 2014). Clusters with the same color code are not significantly different at the 95% confidence level. Black lines within the boxes mark the median and stars represent the mean.