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1 **Hg concentrations and stable isotopes variations in tropical fish species of a**
2 **gold-mining impacted watershed in French Guiana**

3

4 Laure Laffont^{1,*}, Johanna Menges², Sylvaine Goix^{1,3}, Sophie Gentès⁴, Régine Maury-Brachet⁴, Jeroen
5 E. Sonke¹, Alexia Legeay⁴, Patrice Gonzalez⁴, Raphaëlle Rinaldo⁵, Laurence Maurice^{1,*}

6

7 1 – Geosciences Environnement Toulouse, CNRS/IRD/CNES/Université Paul Sabatier, 14 avenue
8 Edouard Belin, 31400 Toulouse, France

9 2 – Section 4.6, Geomorphology, GFZ–German Research Centre for Geosciences, 14473 Potsdam,
10 Germany

11 3 – University Paul Sabatier, 14 avenue Edouard Belin, 31400 Toulouse, France

12 4 – EPOC, EPHE, Univ. Bordeaux, CNRS, 33120 Arcachon, France

13 5 – Parc Amazonien de Guyane, 1 rue Lederson 97354 Remire-Montjoly, France

14

15 *Corresponding authors: laure.laffont@get.omp.eu; laurence.maurice@ird.fr

16

17 **ORCID numbers:** 0000-0001-5850-7937 (L. Laffont), 0000-0001-7223-4787 (J. Menges), 0000-
18 0002-5392-793X (S. Gentes), 0000-0001-7146-3035 (J. Sonke), 0000-0002-1052-142X (A. Legeay),
19 0000-0003-2628-2993 (P. Gonzalez), 0000-0003-3482-3892 (L. Maurice)

20

21 **Key words:** mercury, gold-mining, French Guiana, fish, stable isotopes, methylmercury

22

23 **Abstract:**

24 The aim of the study was to determine if gold mining activities could impact the mercury (Hg)
25 concentrations and isotopic signatures in freshwater fish consumed by riparian people in French
26 Guiana. Total Hg, MeHg concentrations and Hg stable isotopes ratios were analyzed in fish muscles
27 from different species belonging to three feeding patterns (herbivorous, periphytophagous and

28 piscivorous). We compared tributaries impacted by gold-mining activities (Camopi, CR) with a
29 pristine area upstream (Trois-Sauts, TS), along the Oyapock River. We measured $\delta^{15}\text{N}$ and $\delta^{13}\text{C}$ to
30 examine whether Hg patterns are due to differences in trophic level. Differences in $\delta^{15}\text{N}$ and $\delta^{13}\text{C}$
31 values between both studied sites were only observed for periphytophagous fish, due to difference of
32 CN baselines, with enriched values at TS. Total Hg concentrations and Hg stable isotopes
33 fractionations showed that Hg accumulated in fish from both areas have undergone different
34 biogeochemical processes. $\Delta^{199}\text{Hg}$ variation in fish (-0.5 to 0.2‰) was higher than the ecosystem
35 baseline defined by a $\Delta^{199}\text{Hg}$ of -0.66‰ in sediments, and suggested limited aqueous photochemical
36 MeHg degradation. Photochemistry-corrected $\delta^{202}\text{Hg}$ in fish were 0.7‰ higher than the baseline,
37 consistent with biophysical and chemical isotope fractionations in aquatic environment. While THg
38 concentrations in periphytophagous fish were higher in the gold-mining area than in TS, the ensemble
39 of Hg isotope shifts in fish is affected by the difference of biotic (methylation/demethylation) and
40 abiotic (photochemistry) processes between both areas and did therefore not allow to resolve the
41 contribution of gold-mining related liquid Hg(0) in fish tissues. Mercury isotopes of MeHg in fish and
42 lower trophic level organisms can be complementary to light stable isotope tracers.

43

44 **Introduction**

45 Mercury (Hg) is a global pollutant which can induce serious health effects on living organisms
46 depending on its concentration and speciation. The key neurotoxic form of Hg is methylmercury
47 (MeHg) which is produced naturally in aquatic environments by microorganisms (Guimarães et al.,
48 1998; Roulet et al., 2000, 2001; Fleming et al., 2006; Kerin et al., 2006). MeHg is then
49 bioaccumulated and biomagnified along the trophic chain (Mason et al., 1995; Maurice-Bourgoin et
50 al., 1999; Kwon et al., 2012a, Chen et al., 2014) resulting in highly elevated MeHg concentrations in
51 large predator fish (Roulet et al., 2000, 2001, Maury-Brachet et al., 2006). Fish consumption is a
52 major pathway of human exposure to MeHg which can lead to severe health effects such as trembling,
53 eyesight problems, coordination disorders, defects and ultimately death (WHO, 1990). It is therefore
54 important to understand how MeHg is generated and incorporated into the food webs and identify its
55 main sources to the food web.

56 Indigenous riparian communities with a diet heavily relying on fish are especially at risk
57 (Maurice-Bourgoin et al., 1999 and 2000, Mergler et al., 2007). In French Guiana, the regular
58 consumption of local fish leads to elevated Hg concentrations in blood and hair of the local population
59 exceeding the World Health Organization (WHO, 1990) safety limit ($10 \mu\text{g}\cdot\text{g}^{-1}$) (Cordier et al., 1998,
60 Fréry et al., 2001). Neurodevelopmental investigations among MeHg-exposed children in French
61 Guiana also showed significant links between mercury concentrations and the presence of neurological
62 deficiencies (Cordier et al., 2002). The main sources of Hg to the aquatic environment (and ultimately
63 humans) in French Guiana are small-scale gold mining activities and the erosion of tropical soils.
64 Small- and large-scale gold mining activities are the largest Hg emission source (>55%) in South
65 America (Pacyna et al., 2010). The gold mining activities have been greatly enhanced by the increase
66 of the gold price since 1980. Over the last decade gold mining has dramatically impacted the
67 environment by increasing deforestation, soil erosion, surface water turbidity as well as the fishing and
68 hunting pressure (Rimbaud et al., 2017). Large parts of the gold mining operations are clandestine and
69 not controlled by the government. The total gold exploitation in French Guiana has been estimated at 5
70 to 10 tons per year by the WWF (World Wildlife Fund for Nature, 2018). The mean ratio of “liquid
71 Hg used” to “gold produced” is estimated to be 5 (AMAP/UN, 2019) leading to an estimation of 25 to
72 50 tons per year of liquid Hg used for the amalgamation process in this region. Even though the liquid
73 Hg used by small-scale gold-miners is partly recovered by retorting, large quantities of elemental Hg
74 are liberated into the environment, both as vapor emitted to the atmosphere and in its liquid form
75 directly released into soils and sediments (Velásquez-López, P. C., 2010). These locally Hg-
76 contaminated soils and sediments, but also natural and non-gold mining anthropogenic Hg from soils,
77 can then be transferred to the aquatic environment (Meech et al., 1997; Roulet et al., 2000; Dominique
78 et al., 2007a and b; Goix et al., 2019). The speciation processes of liquid Hg were investigated in a
79 freshwater indoor micro-cosmos which revealed Hg transfer between Hg^0 droplets and water column,
80 leading to the formation of aqueous Hg(II) and subsequently MeHg (Dominique et al., 2007a).
81 However, there is no data on natural settings. The direct impact of liquid Hg released during gold
82 mining activities on the aquatic food chain and subsequently on the local population remains few
83 documented (Telmer et al., 2006, Marshall et al., 2018). Soil run-off is an important source of Hg to

84 the hydrosystems. In this region, soils are characterized by elevated organic matter (around $60 \mu\text{g}\cdot\text{g}^{-1}$
85 in the upper horizon in Grimaldi et al., 2008 and Guedron et al., 2006) and clay content and are
86 enriched in Al- and Fe-oxyhydroxydes with a high storage capacity for Hg (Roulet et al., 1998, 2000;
87 Grimaldi et al., 2008; Guedron et al., 2006, 2009). These soils accumulated atmospheric Hg deposition
88 of natural and anthropogenic origin over thousands of years resulting in medium to high Hg
89 concentrations ranging from 0.029 to $0.100 \mu\text{g}\cdot\text{g}^{-1}$ (Goix et al., 2019). Basically, these values are in the
90 low or medium end of the concentration of contaminated soil (Guedron et al., 2006).

91 The variation in the natural abundances of Hg stable isotopes has become a widely used tool
92 during the last few years (Blum, 2011, Sonke and Blum, 2013). Mercury has seven stable isotopes
93 with nominal masses of 196, 198, 199, 200, 201, 202, and 204 and their isotopic variation can be used
94 to identify and trace biogeochemical pathways in analogy to stable isotopes of lighter elements.

95 Hg isotope variations can be explained by mass-dependent fractionation (MDF) and mass-
96 independent fractionation (MIF). MDF is proportional to the mass difference of the Hg isotopes and
97 occurs during redox transformations, biological cycling, and volatilization of Hg (Bergquist and Blum,
98 2009). MIF is predominantly associated with odd numbered Hg isotopes (^{199}Hg and ^{201}Hg) and only
99 induced by a few transformation reactions. The ratio between anomalies $\Delta^{199}\text{Hg}$ and $\Delta^{201}\text{Hg}$ (defined
100 as the difference between measured $\delta^{199}\text{Hg}$ or $\delta^{201}\text{Hg}$ and theoretical $\delta^{199}\text{Hg}$ or $\delta^{201}\text{Hg}$ calculated from
101 mass dependent fractionation only) appears characteristic of two known processes which result in
102 MIF: the nuclear volume effect or the magnetic isotope effect which occurs during photochemical
103 radical pair reactions (Estrade et al., 2009; Bergquist and Blum, 2007). However, in the environment
104 MIF introduced by magnetic isotope effect induced by photochemical reactions is predominant
105 relative to MIF by nuclear volume effect (Blum, 2011, Sonke, 2011). As $\Delta^{199}\text{Hg}$ and $\Delta^{201}\text{Hg}$ signatures
106 remain unchanged during non-photochemical transport or transfer processes, they are especially useful
107 to trace sources, when we know the Hg isotopic signatures, including MIF, of sources, and
108 photochemical transformations of Hg in the environment. More recently, MIF on even isotope ^{200}Hg
109 has been observed and is measured as anomaly $\Delta^{200}\text{Hg}$ (defined as the difference between measured
110 $\delta^{200}\text{Hg}$ and theoretical $\delta^{200}\text{Hg}$ calculated from mass dependent fractionation only). It is used to

111 discriminate between two atmospheric sources: dry and wet deposits. First have negative $\Delta^{200}\text{Hg}$
112 **mostly representative of GEM signature** while **wet deposition** exhibits positive $\Delta^{200}\text{Hg}$ (Gratz et al.,
113 2010; Demers et al., 2013 and 2015; Lepak et al., 2015; Yin et al., 2016; Fu et al., 2016 and Obrist et
114 al., 2017).

115 MDF with a large range of $\delta^{202}\text{Hg}$ is observed in natural samples and in many natural samples
116 anomalies signatures have also been reported (Blum et al., 2014). Bergquist and Blum (2007)
117 observed large positive odd-isotopes anomalies during the experimental photochemical degradation of
118 inorganic Hg (IHg) and MeHg as well as in natural fish samples. They ascribed the fractionation
119 during the photochemical reaction and suggested that the complementary odd-isotopes signatures of
120 the residual MeHg in water gets conservatively incorporated into the food web and can therefore be
121 observed in the fish samples. As explained by Tsui et al., odd-isotopes anomalies are likely
122 attributable to differences in diet or % MeHg (Tsui et al., 2012). In a variety of other studies, no
123 indication of *in vivo* MIF was observed (Gantner et al., 2009; Perrot et al., 2010, 2012; Senn et al.,
124 2010). Hg isotopes have been analyzed in high-trophic level biota to reflect the Hg isotope values of
125 MeHg in the ambient environment as trophic fractionation of Hg isotopes in the lower trophic levels is
126 not significant for Hg(II) and MeHg (Tsui et al., 2020). Kwon et al. (2012b) conducted two controlled
127 experiments to evaluate MDF and MIF during trophic transfer into fish. They found no indication of
128 isotopic fractionation and suggested that stable Hg isotope ratios in fish can be used to trace
129 environmental sources of Hg in aquatic ecosystems. More recently, in Tsui et al. review (2020),
130 authors suggested that Hg isotopes signature, especially odd-MIF, can increase with river size due to
131 reduction in canopy cover which induces increase of photochemistry of Hg and MeHg (Tsui et al.,
132 2013).

133 It is the case of areas impacted by goldmining activities, miners use liquid elemental mercury
134 for the gold amalgamation with a known isotopic signature of mercury (Laffont et al., 2011; Goix et
135 al., 2019). This includes Hg(0) vapor produced by amalgam burning which could be transported in the
136 atmosphere, reduced and redeposited with the precipitation in the forest (foliage, soils, etc.). Actually,
137 Grimaldi et al. 2015 showed that redeposition of Hg(0) vapor after amalgam burning in upper tropical

138 soils is minor relative to natural Hg. However, natural mercury from pristine sediments showed a
139 different isotopic signature than in gold-mined area (Goix et al., 2019). Mercury inputs in sediments as
140 well as in water column, from natural and from goldmining activities, undergo physico-bio-chemical
141 processes in the hydrosystem (desorption/sorption of mercury between liquid and solid phase,
142 absorption/methylation/demethylation in aquatic organisms, and photochemistry in surface water) that
143 induce isotopes fractionation which are highly dependent of the environment; for example, the MIF
144 signature in macroinvertebrates and fish is mediated by the effect of canopy on in-stream
145 photodemethylation (Tsui et al., 2013). The knowledge of the fractionations induced by each of these
146 processes can help to discriminate between the different sources of mercury for identical
147 environmental conditions.

148 To understand the feeding relationships among consumers in a food web, carbon and nitrogen
149 stable isotopes analysis are recognized as relevant tools (Layman et al. 2012). Indeed, nitrogen
150 incorporation by organisms leads to an enrichment in $\delta^{15}\text{N}$ of approximately 3 to 5‰ between each
151 trophic level (Cabana & Rasmussen, 1994; Zanden et al. 2001; Caut et al. 2009), allowing the trophic
152 position of species to be inferred. In contrast, the relatively low enrichment of $\delta^{13}\text{C}$ along food chains
153 (0 to 1‰ between two trophic levels) enables discriminating the different sources of organic carbon
154 (Hobson et al. 2002. Neres-Lima et al. 2017).

155 The main objective of the present study was to determine if gold mining activities impact the
156 Hg concentrations and isotopic signatures of tropical fish species. We studied the Oyapock river basin
157 in French Guiana characterized by a pristine (Trois-Sauts, TS) and a gold mining (Camopi River, CR)
158 area. Specific objectives were: i) to better understand the underlying processes of Hg stable isotopes
159 fractionation in fish species from three different feeding patterns and ii) to assess if Hg isotopes
160 signatures can be used to distinguish anthropogenic from natural Hg in fish muscle, mainly in biofilm-
161 eating fish species. In the frame of the same project (RIMNES research program), Goix et al. (2019)
162 analyzed sediments sampled at the same time and in the same locations than the fish results presented
163 in this article. Our team showed that Hg isotopes distinguish both sources of Hg in sediments and

164 revealed that release of liquid mercury from gold-mining activities directly impact locally exploited
165 rivers.

166 Fish samples with different feeding ecology (herbivorous, periphytophagous and piscivorous)
167 have been analyzed for total Hg and nitrogen ($\delta^{15}\text{N}$) and carbon ($\delta^{13}\text{C}$) stable isotopes, while MeHg
168 concentrations have been determined in periphytophagous fish and Hg stable isotopes have been
169 measured in periphytophagous and piscivorous fish species. Samples were collected in two sub-basins
170 of the Oyapock River, in French Guiana: the Oyapock river upstream at Trois-Sauts village, and the
171 Camopi river, respectively in pristine and gold-mining areas.

172 **Methods**

173 **2.1 Study area**

174 Our studied area is located in the Oyapock basin at the eastern part of French Guiana, at the
175 Brazilian border (Figure 1). The Oyapock River (404 km long) originates in the Tumuk Humak
176 Mountains and runs northward towards the Atlantic Ocean. Its mean annual discharge reaches 800 m^3
177 s^{-1} at the hydrological gauging station close to Saint Georges. Samples were collected 50 km upstream
178 at Camopi village and 250 km upstream at Trois Sauts villages. At the Camopi village, the Camopi
179 River (secondary stream), one of its main tributaries, joins the Oyapock River (primary stream). The
180 Camopi River basin is affected by artisanal and small-scale gold mining activities (ASGM) since the
181 19th century. Nowadays a recent rise in ASGM has been reported on small creeks upstream (tertiary
182 streams), in the Camopi River basin, and on the Oyapock River basin downstream of the confluence
183 with the Camopi River (Parc Amazonien de Guyane, PAG 2017; Rahm et al. 2017). The pristine
184 region of Trois Sauts lies on the Oyapock River 110 km upstream of the Camopi confluence and no
185 gold mining activities have been reported in this area (Gentès et al. 2019).

186 **2.2 Sampling**

187 Fish samples were collected in the Oyapock watershed during the low water stage, in the
188 Camopi river in October 2013 (Hg-contaminated sites impacted by gold mining activities) and in the
189 Oyapock river upstream in October 2012 (the pristine area of Trois Sauts), with the help of local

190 fishermen. Different fishing techniques were used such as nets and fish lines. Following the protocol
191 of Maury-Brachet et al. (2006), fish were dissected on site with a clean stainless steel scalpel and
192 Teflon dissecting forceps and flesh samples were collected. The standard length (cm) and total fresh
193 weight (g) of each individual were measured immediately. Fish samples were frozen in the field using
194 a generator-powered freezer and shipped to GET and EPOC laboratories under dry ice, with final
195 storage in the laboratory at -80°C. Before further analyses, samples were freeze-dried and manually
196 ground to homogenize in an agate mortar.

197 For the purpose of this study, two piscivorous (*Hoplias aimara* and *Boulengerella cuvieri*),
198 two periphytophagous (*Pseudoancistrus barbatus* and *Hypostomus gymnorhynchus*) and two
199 herbivorous (*Myloplus rubripinnis* and *Myloplus ternetzi*) species were selected because 1) they are
200 well represented in the Amazon basin (Le Bail et al. 2012); 2) they are regularly consumed by the
201 local population (Fréry et al. 2001) and therefore represent the main Hg exposure pathway; 3) they
202 represent different trophic levels and have a well-known and specific diet (Keith et al., 2000, Le Bail
203 et al., 2000; Durrieu et al., 2005, Maury-Brachet et al., 2006).

204 The feeding ecology of both piscivorous fish species are mainly fish of varying size, giving
205 them a high trophic position in the food web (see trophic position calculation in results part).
206 Moreover, *Hoplias aimara* was proposed as a bioindicator species for Hg (Durrieu et al., 2005; Gentès
207 et al. 2019). Piscivorous fish were selected by length. *H. aimara* specimen with standard length less
208 than 38 cm could be mistaken with another species (*H. malabaricus*). Small *B. cuvieri* individuals
209 (standard length < 40 cm) are juveniles and their diet is different from that of adult fish. Consequently,
210 we selected *H. aimara* > 38 cm and *B. cuvieri* > 40 cm for analysis.

211 Periphytophagous fish species are benthic primary consumers and consume exclusively
212 periphyton or biofilms on hard substrates (like rocks or immersed tree trunks, etc.). Periphyton is an
213 important food source for the food web, even when its biomass is low (Lamberti, 1996). Moreover,
214 periphyton has been identified as the principal entry point for methylmercury in an Amazonian region
215 (Roulet et al., 2000). Indeed, several studies have shown that periphyton supports high Hg methylation

216 rates due to microbial activity in tropical ecosystems (Guimarães et al., 1998; Acha et al., 2005, Klaus
217 et al. 2016).

218 The both chosen herbivorous species feed exclusively on terrestrial materials from the river
219 banks (leaves, flowers, fruits) and provide information about pelagic primary consumers (Planquette et
220 al., 1996).

221 **2.4 Chemical analysis**

222 **2.4.1 Nitrogen and carbon stable isotopes analysis**

223 We measured nitrogen and carbon ratios in muscles of herbivorous, periphytophagous and
224 piscivorous fish species. All samples were dried and reduced to powder and 0.5 mg were weighed and
225 placed in tin capsules (purity 99.998%) and then analyzed using a continuous flow isotope ratio mass
226 spectrometer in Toulouse (Elemental Microanalysis, UK) in Ecolab laboratory (France). The Isoprime
227 100 spectrometer is coupled in continuous flow mode using pure helium as gas vector to an elemental
228 analyzer Micro Vario for solids combustion. A dual inlet injection system introduces the reference
229 gases (N₂ and CO₂).

230 The ¹³C/¹²C (denoted δ¹³C) and ¹⁵N/¹⁴N (denoted δ¹⁵N) ratios, expressed in ‰, were calculated
231 as the relative differences between the sample and the conventional standard following Peterson and
232 Fry (1987):

$$233 \quad \delta^{15}\text{N} \text{ or } \delta^{13}\text{C} (\text{‰}) = [(R_{\text{sample}} - R_{\text{standard}}) / R_{\text{standard}}] \times 1,000 \text{ (Eq. 1)}$$

234 where R is ¹³C/¹²C or ¹⁵N/¹⁴N (Peterson and Fry, 1987). The international reference material standard
235 is atmospheric N₂ for nitrogen and Vienna Pee Dee Belemnite for carbon.

236 Standards were run in duplicate every twenty measurements. Three internal standards of urea,
237 alanine and acetanilid were analyzed. The obtained average values were δ¹⁵N = -0.45 ± 0.14 ‰ and
238 δ¹³C = -36.33 ± 0.26 ‰ (n = 9, 2SD) for urea, δ¹⁵N = 8.38 ± 0.23 ‰ and δ¹³C = -23.03 ± 0.18 ‰ (n =
239 6, 2SD) for alanine and δ¹⁵N = -3.89 ± 0.46 ‰ and δ¹³C = -33.29 ± 0.14 ‰ (n = 6, 2SD) for acetanilid.
240 The highest analytic precisions were 0.46 ‰ for δ¹⁵N and 0.26 ‰ for δ¹³C (2SD).

241 Measured C/N ratios provide an indication of the fat content in the tissue analyzed (Post et al.
242 2007). Because lipids are ^{13}C -depleted relative to proteins, high lipid content (with $\text{C/N} > 4$) in
243 organisms affects measured $\delta^{13}\text{C}$ ratios. In our study, all organisms had a $\text{C/N} < 4$, thus not requiring
244 lipid extraction.

245 **2.4.2 Total Hg concentrations**

246 Fish samples were immediately stored in sealed polyethylene bags and frozen at $-18\text{ }^{\circ}\text{C}$. Back
247 to the laboratories, they were freeze-dried and analyzed a couple of months after sampling. Total Hg
248 concentrations (THg) were measured on 108 fish muscle samples, using an Advanced Mercury
249 Analyzer (AMA254) (GET laboratory). Each sample was combusted inside the AMA at 750°C for
250 150 seconds. The released Hg was trapped in a gold trap and subsequently released and measured with
251 an integrated atomic absorption spectrometer. The accuracy of the measurements was assigned by the
252 analysis of certified reference materials ERM-CE464 (tuna fish) and DOLT4 (dogfish liver) (for the
253 piscivorous fish species) as well as TORT 2 (lobster hepatopancreas) for the less concentrated
254 periphytophagous fish species.

255 The measured concentrations for the reference materials were in agreements with the certified
256 values for TORT2 (THg = $0.307 \pm 0.004\ \mu\text{g}\cdot\text{g}^{-1}$; $n = 6$; **recovery: 114%**) and DOLT4 (THg = $2.385 \pm$
257 **$0.003\ \mu\text{g}\cdot\text{g}^{-1}$** ; $n = 4$; **recovery : 92%**). For CE464 the recovery was $91 \pm 1\%$ ($4.757 \pm 0.208\ \mu\text{g}\cdot\text{g}^{-1}$, $n =$
258 3). Natural samples were all measured in duplicate. If the difference between the duplicate
259 measurements was above 10%, a third measurement was carried out. All THg concentrations are
260 reported on a dry weight basis.

261 **2.4.3 Hg speciation analyses**

262 We assumed that %MeHg in piscivorous fish is close to 100% (Maury-Brachet et al. 2006)
263 and chose to not analyze mercury speciation of these samples. However, a number of nineteen muscle
264 samples of periphytophagous fish and two certified reference materials, TORT2 and NIST2976, were
265 analyzed. Speciation analyses have been made using double-spike species specific isotope dilution
266 method with GC-ICP-MS detection at the GET laboratory (Monperrus et al. 2008). Subsamples of 200
267 mg were digested with 5 mL of TMAH in a microwave Discover SP-D (CEM society) with a ramp of

268 2 min and constant heating during 4 min at 70°C and stirring (Navarro et al. 2011). After cooling, an
269 aliquot of the extract was transferred to glass vials and 5 ml HAc/NaAc buffer solution (pH 3.9) was
270 added to the aliquot. Then, an appropriate amount of isotopically enriched ¹⁹⁹IHg and ²⁰¹MeHg was
271 added to each sample (Monperrus et al, 2008). After letting the sample equilibrate for two hours, the
272 pH was adjusted to pH 3.9 using bi-distilled HCl. The species were then propylated by adding 0.2 ml
273 of newly prepared Sodium-tetra(n-propyl)borate (NaBPr₄) 10% (w/v). Then, 2 ml of hexane was
274 added and each vial was manually shaken for 5 min to extract all the Hg species into the organic
275 phase. An aliquot of the organic phase was then transferred into a GC-vial and kept frozen until
276 analysis. The samples were analyzed using a GC (Thermo) coupled to a sector field ICP-MS (Thermo
277 Element XR) at the GET laboratory. The measured yield of MeHg for the two reference materials, (58
278 % for TORT2 and 49 % for NIST2976), are consistent with the certified values of 56 % and 46 %,
279 respectively. The average yield of the analysis was 98±8 % quantified by comparison of the sum of the
280 MeHg and IHg concentrations measured with the described method and THg analysis with AMA254.

281 **2.4.4 Hg stable isotope analyses**

282 Appropriate amounts of samples (0.5 g of periphytophagous fish muscle and 0.05 g of
283 piscivorous fish muscle) were digested in 5 ml inverse aqua regia (HNO₃ and HCl, 3:1 v:v) at 120°C
284 for 4 h in closed glass vials on a heating plate. After cooling, 1 ml of H₂O₂ was added and samples
285 were heated again at 120°C for at least 4 h. The average yield of the digestion was 103±6 % for
286 periphytophagous and 104±5 % for piscivorous fish samples quantified by comparison of the results
287 from the MC-ICP-MS and THg analysis with AMA254. In 2013-2014, our technical materials did not
288 allow us to analyze Hg isotopes of herbivorous fish due to low THg concentration.

289 Hg stable isotope ratios were measured by continuous flow cold vapor generation (using
290 Sn(II) reduction) multi collector inductively coupled plasma mass spectrometry (CV-MC-ICPMS,
291 Thermo Finnigan Neptune, Germany) at the GET laboratory (Toulouse, France) according to
292 previously published methods (Laffont et al., 2009, 2011; Masbou et al., 2013, 2014; Enrico et al.,
293 2016). Instrumental mass bias was corrected by sample bracketing using the international Hg standard
294 NIST SRM 3133 at matching concentrations (several sequences between 0.8 and 2.5 ppb depending

295 on initial THg of fish) and prepared in the same matrix than samples. Hg isotopic composition is
 296 reported as δ -values, which represents the deviation from the bracketing standard and expressed in
 297 permil (‰) using the following equation:

$$298 \quad \delta^{xxx}\text{Hg} = \left(\frac{\left(\frac{xxx\text{Hg}}{198\text{Hg}} \right)_{\text{sample}}}{\left(\frac{xxx\text{Hg}}{198\text{Hg}} \right)_{\text{SRM3133}}} - 1 \right) * 1000 \text{ (Eq. 2)}$$

299 MIF is quantified as the δ -value deviation from the theoretical MDF using the following equation:

$$300 \quad \Delta^{xxx}\text{Hg} = \delta^{xxx}\text{Hg}_{\text{sample}} - \beta * \delta^{202}\text{Hg}_{\text{sample}} \text{ (Eq. 3)}$$

301 Where β -values for isotopes ^{199}Hg , ^{200}Hg , ^{201}Hg and ^{204}Hg are 0.252, 0.502, 0.752, and 1.493
 302 respectively according to the kinetic MDF law (Blum and Bergquist, 2007). Long-term reproducibility
 303 of Hg isotope measurements was assessed by analyzing two reference materials UM-Almaden and
 304 ETH in-house Fluka along with the samples at the same THg concentrations. The obtained values for
 305 UM-Almaden are $\delta^{202}\text{Hg} = -0.52 \pm 0.14$ ‰, $\Delta^{199}\text{Hg} = -0.03 \pm 0.08$ ‰ and $\Delta^{200}\text{Hg} = 0.03 \pm 0.04$ ‰
 306 (mean \pm 2SD, n = 7) and for ETH in-house Fluka $\delta^{202}\text{Hg} = -1.53 \pm 0.14$ ‰, $\Delta^{199}\text{Hg} = -0.10 \pm 0.04$ and
 307 $\Delta^{200}\text{Hg} = 0.02 \pm 0.04$ ‰ (mean \pm 2SD, n = 14) and agree with previously published values (Blum and
 308 Bergquist, 2007; Smith et al., 2014). The certified reference materials TORT2 and ERM-CE464 were
 309 also analyzed. Measured isotopic compositions for reference materials are consistent with previously
 310 published values (Laffont et al., 2009; Masbou et al., 2013). Uncertainties applied to samples were the
 311 larger of either the 2SD on the ETH in-house Fluka or the 2SD on sample replicate when available and
 312 larger than the 2SD of ETH in-house Fluka.

313 **2.5 Data analysis**

314 **2.5.1 Trophic position calculation for piscivorous fish**

315 The position of an organism within a food chain can be deduced from the relatively constant $\delta^{15}\text{N}$
 316 enrichment observed between each trophic level (Vander Zanden and Rasmussen, 1996). The trophic
 317 position (TP) of piscivorous fish was calculated based on $\delta^{15}\text{N}$ values and following the equation of
 318 Bergamino et al. (2011):

$$319 \quad \text{TP}_i = [(\delta^{15}\text{N}_i - \delta^{15}\text{N}_{\text{pc}}) / 3.4] + 2 \text{ (Eq 4)}$$

320 where TP_i represents the average position of species i ; $\delta^{15}N_i$ the average $\delta^{15}N$ value of species i ; $\delta^{15}N_{pc}$
321 the average $\delta^{15}N$ value of primary consumers; 3.4 the mean $\delta^{15}N$ trophic enrichment occurring per
322 trophic level (Post, 2002); and 2, the trophic position of the baseline organism (= primary consumer,
323 here herbivorous fish species were selected because they present the lowest $\delta^{15}N$ values, in comparison
324 to periphytophagous fish, see result part 3.1).

325 **2.5.2 Statistical analysis**

326 Statistical analyses were done using ©R (Version 3.4.4). Comparison tests were conducted
327 with Wilcoxon (medians) as distribution is not normal and sample size low. A confidence level of 0.95
328 was chosen. Means or medians were considered to be significantly different when the p value was less
329 than 0.05.

330 **Results**

331 These are first data of THg, MeHg concentrations and stable Hg isotopes ratios for different
332 fish species highly consumed in French Guiana by local population. All the results are detailed in the
333 Supplementary information (Table S1).

334

335 **3.1 Nitrogen and carbon stable isotopes ratios in fish**

336 Carbon ($\delta^{13}C$) and nitrogen ($\delta^{15}N$) stable isotopes individual values for herbivorous,
337 periphytophagous and piscivorous fish from both sites are plotted in Figure 2. Results shown that the
338 $\delta^{15}N$ is similar between two sites for piscivorous fish (Wilcoxon test, $W = 26$, $p > 0.05$), but not for
339 periphytophagous fish which it is significantly different (Wilcoxon test, $W = 62$, $p = 0.013$) with
340 higher values in TS (10.8 ± 0.6 ‰, $n = 12$) than in CR (9.8 ± 0.7 ‰, $n = 6$). We cannot calculate a
341 Wilcoxon test for herbivorous fish as $n = 5$ at CR but the average $\delta^{15}N$ is close for both site (6.2 ± 0.3
342 ‰, $n = 9$, at TS and 6.2 ± 0.5 ‰, $n = 5$, at CR). Trophic positions (TP) were 3.8 ± 0.1 and 3.9 ± 0.1 for
343 piscivorous fish for CR and TS, respectively and were not statistically different between both sites
344 (Wilcoxon test, $W = 0.94$, $p = 0.34$).

345 Periphytophagous fish present a variation of $\delta^{13}C$ (Fig. 2) ranging from -30.5 to -25.8 ‰ with an
346 average of -28.3 ± 1.3 ‰ ($n = 12$) at TS and from -24.0 to -27.9 ‰ with an average of -26.0 ± 1.3 ‰ ($n =$

347 6) at CR, showing a slightly more positive $\delta^{13}\text{C}$ at CR than TS (Wilcoxon test, $W = 7$, $p = 0.005$). The
348 $\delta^{13}\text{C}$ values of herbivorous fish (-27.2 ± 0.2 ‰, $n = 9$, at TS and -26.7 ± 0.3 ‰, $n = 5$, at CR) are very
349 close between both sites (Wilcoxon test not possible with $n = 5$). Piscivorous fish $\delta^{13}\text{C}$ values are also
350 statistically similar between both sites: -26.2 ± 0.5 ‰ ($n = 6$) at TS and -26.2 ± 0.2 ‰ ($n = 6$) at CR
351 (Wilcoxon test, $W = 19$, $p = 0.94$).

352 **3.2 Total mercury concentrations in fish**

353 THg concentrations (Fig. 3) in herbivorous species ($n = 36$) vary between 0.006 and 0.037
354 $\mu\text{g.g}^{-1}$ dw and are significantly higher at TS compared to CR (0.022 ± 0.006 $\mu\text{g.g}^{-1}$ at TS and
355 0.012 ± 0.008 $\mu\text{g.g}^{-1}$ at CR, Wilcoxon test, $W = 276$, $p = 8.1 \times 10^{-5}$). In periphytophagous species ($n = 41$),
356 THg concentrations vary between 0.017 and 0.248 $\mu\text{g.g}^{-1}$ dw and are significantly higher at CR
357 relatively to TS (0.13 ± 0.05 $\mu\text{g.g}^{-1}$ at TS and 0.20 ± 0.04 $\mu\text{g.g}^{-1}$ at CR, Wilcoxon test, $W = 28$, $p =$
358 0.003). In piscivorous species ($n = 30$), THg concentrations range from 0.216 to 6.346 $\mu\text{g.g}^{-1}$ dw and
359 are not significantly different between CR and TS (average of 2.0 ± 0.9 $\mu\text{g.g}^{-1}$ at TS and 2.9 ± 1.5 $\mu\text{g.g}^{-1}$
360 at CR, Wilcoxon test, $W = 65$, $p = 0.094$). THg concentrations in all piscivorous fish samples (except
361 one) exceeded the US EPA (2012) recommendation THg value of 0.256 $\mu\text{g.g}^{-1}$ dw for a daily
362 consumption of 0.113 kg (ww)/day of fish for a pregnant women of 78 kg.

363 **3.3 Methylmercury concentrations in fish**

364 The %MeHg has only been measured in periphytophagous species because it reaches more
365 than 95 % in piscivorous study species (Maury-Brachet et al., 2006): it varies between 92 and 95 %
366 with a mean of 95 ± 1 % (1SD, $n = 6$) in CR and between 82 and 95 % with a mean of 89 ± 5 % (1SD, n
367 $= 13$) in TS (Table S1). The difference of %MeHg between both locations is significant (Wilcoxon
368 test, $W = 4$, $p = 0.0009$) with the %MeHg higher at CR. Moreover, we observe that the variance of
369 %MeHg is higher at TS than at CR (20.9 and 1.3 respectively, variance unity is the square of %).

370 **3.4 Mercury isotopes signatures**

371 For periphytophagous fish, $\delta^{202}\text{Hg}$ values vary between -1.40 ‰ and -0.89 ‰ (mean of $-$
372 1.14 ± 0.16 ‰ (1SD)) at TS and between -1.41 ‰ and -1.03 ‰ (mean of -1.21 ± 0.14 ‰) at CR. For

373 piscivorous fish, $\delta^{202}\text{Hg}$ values vary between -1.68 ‰ and -1.17 ‰ (mean of -1.42 ± 0.11 ‰) at TS and
374 between -1.68 ‰ and -0.95 ‰ (mean of -1.29 ± 0.21 ‰) at CR (Fig. 4). Regarding the stable Hg
375 isotopes anomalies, $\Delta^{199}\text{Hg}$ values for periphytophagous vary between -0.49 ‰ and 0.00 ‰ (mean of -
376 0.30 ± 0.16 ‰) at TS and between -0.60 ‰ and 0.20 ‰ (mean of 0.06 ± 0.12 ‰) at CR. $\Delta^{199}\text{Hg}$ values
377 for piscivorous vary between -0.50 ‰ and -0.03 ‰ (mean of -0.38 ± 0.11 ‰) at TS and between -0.26
378 ‰ and -0.06 ‰ (mean of -0.20 ± 0.06 ‰) at CR (Fig. 4). Even isotope anomaly for periphytophagous
379 fish, $\Delta^{200}\text{Hg}$, varies between -0.09 ‰ and -0.04 ‰ (mean of -0.06 ± 0.02 ‰) at TS and between -0.09
380 ‰ and -0.01 ‰ (mean of -0.05 ± 0.03 ‰) at CR. $\Delta^{200}\text{Hg}$ values for piscivorous vary between -0.08 ‰
381 and -0.04 ‰ (mean of -0.06 ± 0.01 ‰) at TS and between -0.07 ‰ and -0.02 ‰ (mean of -0.04 ± 0.02
382 ‰) at CR (Table S1).

383 Despite the lower number of samples at CR, we investigate the $\Delta^{199}\text{Hg}/\Delta^{201}\text{Hg}$ regression
384 slope for each fish feeding pattern in both pristine and gold-mining areas. Slopes of the regression
385 lines are 1.31 ± 0.06 ($r^2 = 0.96$, $n = 16$, 1SD) and 1.37 ± 0.09 ($r^2 = 0.95$, $n = 15$, 1SD) for
386 periphytophagous and piscivorous respectively at TS. In the gold-mining area of CR, the regression
387 line slopes are 1.61 ± 0.34 ($r^2 = 0.85$, $n = 6$, 1SD) and 0.84 ± 0.12 ($r^2 = 0.86$, $n = 10$, 1SD) for
388 periphytophagous and piscivorous fish respectively. Slopes for periphytophagous fish between TS and
389 CR are not significantly different (ANCOVA test, $p = 0.22$) while slopes for piscivorous are
390 significantly different (ANCOVA test, $p = 0.002$).

391 Discussion

392 4.1 $\delta^{15}\text{N}$ and $\delta^{13}\text{C}$ differences between both study locations

393 The significantly higher $\delta^{15}\text{N}$ values measured in periphytophagous fish from TS compared to
394 CR could be explained by difference in the composition of biofilm between both locations. The
395 significantly higher (but slight from an ecological point of view) $\delta^{15}\text{N}$ values measured in
396 periphytophagous fish from TS compared to CR could be explained by several environmental
397 parameters specific to each site. It could be due, for example, to the difference in the composition and
398 age of the biofilm between both locations (chlorophyll a and a/b ratio in periphyton), A variation in
399 periphyton composition could be explained by a difference in transparency of water due to the

400 occurrence of suspended particles (which could be linked to the gold mining activity or the soil use) or
401 to the difference in canopy cover over the river (Oyapock river at TS receives less light because of a
402 higher vegetation cover than the Camopi river at the sampling sites). Another explanation for the
403 difference in $\delta^{15}\text{N}$ measured values in periphytophagous fish between both sites could be linked to the
404 difference in food resources accessibility which may be due to the variation in the vegetation density
405 or in the grazing intensity but also to the different predation pressure on these fish species.

406 Goix et al. (2019) observed that sediments in TS contained more organic carbon but also more
407 MeHg (in %) than in CR. On the contrary, we observed that %MeHg in periphytophagous fish species
408 is generally lower but more variable at TS than at CR. This suggests that there is not direct exposure
409 route from methylmercury contained in sediments, and that periphytophagous fish contamination is
410 mainly due to trophic transfer. Moreover, the large variation of $\delta^{13}\text{C}$ values of periphytophagous fish
411 at both locations suggests multiple C sources which will need to be investigated.

412

413 **4.2 Total mercury concentrations in fish**

414 Average total Hg concentration in fish muscles increased according to the trophic levels. In
415 herbivorous fish species studied, THg concentrations are 125 times lower compared with piscivorous
416 fish species levels. Indeed, herbivorous fish eat terrestrial plants (Planquette et al. 1996) which contain
417 low levels of Hg and MeHg. Terrestrial plants absorb Hg from substrate soil (via the roots) and the
418 atmosphere (via the leaves) which are reservoirs of mainly inorganic Hg species. THg concentrations
419 in the herbivorous fish studied are, thus, much lower than herbivorous fish eating aquatic plants
420 species (Roulet and Maury-Brachet, 2001).

421 Periphytophagous fish have higher THg concentrations at CR than at TS. As explained in the
422 methods section, periphytophagous fish ingest a mix of bacteria, algae and fungi contained in the
423 biofilm. Inorganic Hg in the biofilm is methylated into MeHg subsequently absorbed and
424 bioaccumulated by periphytophagous fish (Guimarães et al., 1998; Acha et al. 2005). The higher THg
425 concentration measured in the periphytophagous fish could result from higher MeHg concentrations in
426 biofilms at CR (Le Bail et al., 2000). In our previous study, we measured higher THg concentration in

427 sediments at CR than at TS. We hypothesize that this THg is first desorbed from sediments and then
428 reached the water column in dissolved form, methylated into the biofilm after its sorption and then
429 ingested by fish (Laperche et al., 2007, Goix et al., 2019).

430 Piscivorous fish had the same THg concentrations at CR than at TS (Wilcoxon, $W = 65$, $p =$
431 0.094), while CR is greatly Hg-impacted by goldmining activities. This could not be explained by
432 different size ranges of fish samples (average of 532 ± 103 cm ($n = 19$) and 490 ± 68 cm ($n = 11$) at TS
433 and CR, respectively), nor to differences in their trophic position between both sites. However, the
434 distribution of prey fish depends on several factors such as hydrological characteristics (river size,
435 turbidity and flow, season, etc.), local ecological factors and variability, and fisheries or gold-mining
436 pressures (Maury-Brachet et al., 2020).

437 **4.3 Hg isotope signatures in relation to the feeding pattern of the fish species**

438 Very few Hg isotopic studies on river fish flesh have been published. In the Bolivian Amazon,
439 Laffont et al. (2009) analyzed fish from the Beni river and its floodplain lakes. Three other studies
440 have been conducted on river fish by Janssen et al. (2019) in Northeastern USA streams, Donovan et
441 al. (2016) in the Yuba river (California, USA) and Tsui et al. (2012) in the Eel river (California, USA).

442 The mass dependent isotopic signature, $\delta^{202}\text{Hg}$, for all fish from this study varies between -
443 1.68 ‰ and -0.89 ‰. This signature is more negative than the ones observed for river fish in literature
444 including the one in the Bolivian Amazon basin. However, $\delta^{202}\text{Hg}$ in sediments is also more negative
445 than the ones in the literature cited in the precedent paragraph. The average Hg isotopic signature from
446 pristine sediments of the Oyapock R. (Goix et al., 2019) were $\delta^{202}\text{Hg} = -2.27$ ‰, $\Delta^{199}\text{Hg} = -0.66$ ‰
447 and $\Delta^{200}\text{Hg} = -0.04$ ‰ ($n = 27$). Negative $\delta^{202}\text{Hg}$, $\Delta^{199}\text{Hg}$ and $\Delta^{200}\text{Hg}$ of pristine sediments and soils
448 reflected dry deposition of atmospheric mercury mostly after vegetation uptake by plants and
449 subsequent transfer to the river system (Goix et al., 2019). The MIF of even isotopes is commonly
450 associated with either dry or wet atmospheric deposition that display negative and positive $\Delta^{200}\text{Hg}$,
451 respectively (Fu et al., 2016 and Obrist et al., 2017). Average $\Delta^{200}\text{Hg}$ anomaly close to -0.05 ‰
452 measured in fish from the Oyapock basin indicated that Hg sources for fish is atmospheric Hg^0 as
453 observed for sediments. It could be Hg desorbed and dissolved from sediments and also from

454 vegetation (foliage/litter) inputs into the water column (Jiskra et al., 2017; Woerndle et al., 2018).
455 Recent studies monitoring the export of Hg via streamflow in forested ecosystems have also shown
456 that the majority of Hg exported was derived from previously dry deposited Hg in the forested
457 watersheds, as their isotopic signatures resemble those of foliage/litter and forest floor rather than
458 direct precipitation (Tsui et al., 2020).

459 Following Janssen et al. (2019), Tsui et al. (2012) and Donovan et al. (2016), we applied a
460 $\delta^{202}\text{Hg}$ correction to calculate the $\delta^{202}\text{Hg}$ before isotopic fractionation induced by photochemistry. This
461 calculation has been first made by Gehrke et al. (2011) using experimental relationships to subtract
462 the known MDF and MIF that occurs during photochemical degradation and calculating $\Delta^{199}\text{Hg}/\delta^{202}\text{Hg}$
463 slope to obtain an estimated $\delta^{202}\text{Hg}$ value for prephotodegraded MeHg and IHg. Actually, we used
464 Bergquist and Blum (2007) experimental data: photoreduction of mercury with 1 mg/L C (slope 1.15)
465 and photodemethylation of MeHg with 1 mg/L C (slope 2.43) (average 2.7 mg.L⁻¹ (n = 16) of DOC
466 was measured in the study rivers (database of the ANR-RIMNES Research Program)). Assuming that
467 mercury in water column of TS and CR came mainly from sediments, pre-photodegraded mercury has
468 $\Delta^{199}\text{Hg}$ close to the $\Delta^{199}\text{Hg}$ of pristine sediments, the average $\delta^{202}\text{Hg}$ of pre-photodegraded MeHg is
469 estimated at -1.54 ‰ and -1.70 ‰ for piscivorous fish at TS and CR respectively and at -1.29 ‰ and -
470 1.51 ‰ for periphytophagous fish at TS and CR respectively. These values are much higher than the
471 $\delta^{202}\text{Hg}$ in the pristine sediment ($\delta^{202}\text{Hg} = -2.27$ ‰), suggesting there is a positive offset of around +0.7
472 ‰ (apart from periphytophagous fish from TS) between sediments and fish, consistent with Janssen et
473 al. (2019), Gehrke et al. (2011) and Tsui et al. (2012) observations. This offset is the net result of
474 biotic MDF during IHg methylation (biotic methylation, biotic MeHg degradation, etc.).

475 The odd-isotopes anomalies of Hg of periphytophagous fish reflect the isotopic signature of
476 dissolved MeHg. Indeed, the slopes ($\Delta^{199}\text{Hg}/\Delta^{201}\text{Hg}$) close to 1.3 have been interpreted as the result of
477 partly photodegraded MeHg in river water, bioaccumulated by fish (Bergquist and Blum 2007). But
478 the latter slope close to 1 is more likely due to photodegradation of IHg prior to its methylation and
479 ingestion by fish (Bergquist and Blum 2007). The offset of around -0.2 ‰ observed in $\Delta^{199}\text{Hg}$ between
480 piscivorous and periphytophagous fish for each location could indicate that sources of this MeHg are

481 different for the two fish diets: piscivorous fish are opportunistic hunters and integrate the Hg
482 signature of the entire environment while periphytophagous are specific biofilm consumers.

483 Moreover, odd-isotopes anomalies observations point out the fact that both areas have
484 different aquatic processes affecting Hg: $\Delta^{199}\text{Hg}$ is more positive for periphytophagous fish at CR and
485 $\Delta^{199}\text{Hg}/\Delta^{201}\text{Hg}$ regression slope at CR is close to 1 for piscivorous fish. Tsui et al. (2012) observed the
486 same slope for terrestrial predators. According to $\delta^{13}\text{C}$ and $\delta^{15}\text{N}$ values which are similar between two
487 areas for piscivorous fish, we could suppose that their food sources are quite the same. These
488 observations can only be explained by differences in photochemistry processes between both areas
489 induced by a different water composition, sunlight exposure (canopy coverage), river flow, and others
490 parameters of water dynamic and chemistry.

491 Consequently, Hg isotopic signatures in fish are affected by the difference of biotic
492 (methylation/demethylation) and abiotic (photochemistry) processes between both areas. We suppose
493 that both aquatic systems are slightly different as explained before and, moreover, disturbed by gold-
494 mining activities by inputs of suspended particles.

495

496 **Conclusion**

497 In periphytophagous fish species, THg concentrations were higher in the gold-mining (CR) than in the
498 pristine (TS) area whereas $\delta^{13}\text{C}$ values were enriched and $\delta^{15}\text{N}$ depleted in CR compared to TS. As
499 these fish exclusively eat biofilm settled on immersed rocks, adventive roots or plant residues, we
500 hypothesize that this difference is due to a different composition of the biofilm between two sites. We
501 showed that Hg isotopic signatures in fish caught in pristine and gold-mined basins have undergone
502 different speciation processes. Variations in Hg isotopes fractionation are attributed to: i) the
503 difference in biotic processes and MeHg sources shown by variations of $\delta^{202}\text{Hg}$, $\delta^{15}\text{N}$ and $\delta^{13}\text{C}$ in both
504 locations, and ii) variation of abiotic processes such as photochemistry between both areas shown by
505 the difference of odd-isotopes anomalies and $\Delta^{199}\text{Hg}/\Delta^{201}\text{Hg}$ regression slope between fish species and
506 locations.

507 To conclude, Hg isotopic signatures in fish are affected by the difference of biotic
508 (methylation/demethylation) and abiotic (photochemistry) processes between both areas and did
509 therefore not allow to resolve the contribution of gold-mining related liquid Hg(0) in fish tissues.
510 Mercury isotopes of MeHg in fish and lower trophic level organisms can be complementary to light
511 stable isotope tracers.

512

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524

525 **Authors' contribution**

526 LM was the coordinator of the RIMNES Research Program (funded by the French ANR). LM and
527 RM-B designed the study dedicated to the ichthyologic part of the program. Formal analysis (THg,
528 speciation and Hg stable isotopes) and geochemical lab work were performed by LL, JM and SGx
529 under the supervision of JS. RR helped with logistics in French Guiana. Fish sampling was realized by
530 RM-B, AL and PG. Statistical analysis were performed by LL and SGx. Funds were acquired by LM
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536

537 **Data availability:** the online version contains the raw data available in the supplementary material.

538

539 **Compliance with ethical standards**

540

541 **Ethical approval:** Not applicable.

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546

547 **References**

548 Acha D., Iñiguez V., Roulet M., Guimarães J. R. D., Luna R., Alanoca L. and Sanchez S. (2005).
549 Sulfate-reducing bacteria in floating macrophyte rhizospheres from an Amazonian floodplain lake in
550 Bolivia and their association with Hg methylation. *Applied and Environmental Microbiology* 71:
551 7531-7535

552

553 AMAP/UN Environment. (2019). Technical Background Report for the Global Mercury Assessment
554 2018. Arctic Monitoring and Assessment Programme, Oslo, Norway/UN Environment Programme,
555 Chemicals and Health Branch, Geneva, Switzerland.

556

557 Bergamino, L., Lercari, D., Defeo, O. (2011). Food web structure of sandy beaches: Temporal and
558 spatial variation using stable isotope analysis. *Estuar. Coast. Shelf Sci.* 91(4): 536-543

559

560 Bergquist B. A., and Blum J. D. (2007). Mass-Dependent and -Independent Fractionation of Hg
561 Isotopes by Photoreduction in Aquatic Systems. *Science* 318: 417–420

562

563 Bergquist B. A., and Blum J. D. (2009). The Odds and Evens of Mercury Isotopes: Applications of
564 Mass-Dependent and Mass-Independent Isotope Fractionation. *Elements* 5: 353–357

565

566 Blum J. D. (2011). Applications of stable mercury isotopes to biogeochemistry. In *Handbook of*
567 *Environmental Isotope Geochemistry*. Springer 229–245

568

569 Blum J. D. and Bergquist B. A. (2007). Reporting of variations in the natural isotopic composition of
570 mercury. *Anal. Bioanal. Chem.* 388: 353–359

571
572 Blum J. D. Sherman L. S. and Johnson M. W. (2014). Mercury Isotopes in Earth and Environmental
573 Sciences. *Annu. Rev. Earth Planet. Sci.* 42: 249–269
574
575 Cabana G. and Rasmussen, J. B. (1994). Modelling food chain structure and contaminant
576 bioaccumulation using stable nitrogen isotopes. *Nature* 372 (6503): 255.
577
578 Caut S., Angulo E. and Courchamp F. (2009). Variation in discrimination factors ($\Delta^{15}\text{N}$ and
579 $\Delta^{13}\text{C}$): the effect of diet isotopic values and applications for diet reconstruction. *Journal of Applied*
580 *Ecology* 46 (2): 443-453
581
582 Chen C. Y., Borsuk M.E., Bugge D. M., Hollweg T., Balcom P. H., Ward D. M., Williams J. and
583 Mason R. P. (2014). Benthic and Pelagic Pathways of Methylmercury Bioaccumulation in Estuarine
584 Food Webs of the Northeast United States. *PLoS ONE* 9, e89305.
585
586 Cordier S., Grasmick C., Paquier-Passelaigue M., Mandereau L., Weber J.-P. and Jouan M. (1998).
587 Mercury Exposure in French Guiana: Levels and Determinants. *Arch. Environ. Health Int. J.* 53: 299–
588 303
589
590 Cordier S., Garel M., Mandereau L., Morcel H., Doineau P., Gosme-Seguret S., Josse D., White R.
591 and Amiel-Tison C. (2002). Neurodevelopmental Investigations among Methylmercury-Exposed
592 Children in French Guiana. *Environ. Res.* 89: 1–11
593
594 Demers J. D., Blum J. D. and Zak D. R. (2013). Mercury isotopes in a forested ecosystem:
595 Implications for air-surface exchange dynamics and the global mercury cycle. *Global Biogeochem.*
596 *Cy.* 27 (1): 222–238.
597
598 Demers J. D., Sherman L. S., Blum J. D., Marsik F. J. and Dvonch, J. T. (2015). Coupling atmospheric
599 mercury isotope ratios and meteorology to identify sources of mercury impacting a coastal
600 urban/industrial region near Pensacola, Florida, USA. *Global Biogeochem. Cy.* 29 (10): 1689–1705.
601
602 Donovan P., Blum J. D., Bliss Singer M., Marvin-DiPasquale M. and Tsui M. T. K. (2016). Isotopic
603 composition of inorganic mercury and methylmercury downstream of a historical gold-mining region.
604 *Environ. Sci. Technol.* 50: 1691-1702
605
606 Dominique Y., Muresan B., Duran R., Richard S. and Boudou A. (2007). Simulation of the Chemical
607 Fate and Bioavailability of Liquid Elemental Mercury Drops from Gold Mining in Amazonian
608 Freshwater Systems. *Environ. Sci. Technol.* 41: 7322–7329
609
610 Dominique Y., Maury-Brachet R., Muresan B., Vigouroux R., Richard S., Cossa D., Mariotti A. and
611 Boudou A. (2007). Biofilm and mercury availability as key factors for mercury accumulation in fish
612 (*Curimata cyprinoides*) from a disturbed Amazonian freshwater system. *Environmental toxicology and*
613 *chemistry/SETAC* 26: 45–52.
614
615 Dominique Y. (2008). Contamination par les différentes formes chimiques du mercure de la
616 composante biologique du barrage hydroélectrique de Petit-Saut et des zones amont/aval du fleuve
617 Sinnamary, en Guyane française (études in situ et approches expérimentales)
618

619 Durrieu G., Maury-Brachet R. and Boudou A. (2005). Goldmining and mercury contamination of the
620 piscivorous fish *Hoplias aimara* in French Guiana (Amazon basin). *Ecotoxicol. Environ. Saf.* 60: 315–
621 323

622

623 Enrico M., Le Roux G., Maruszczak N., Heimbürger L-E., Claustres A., Fu X., Sun R. and Sonke J. E.
624 (2016). Atmospheric mercury transfer to peat bogs dominated by gaseous elemental mercury dry
625 deposition. *Environ. Sci. Technol.* 50: 2405–2412

626

627 Estrade N., Carignan J., Sonke J. E. and Donard, O. F. X. (2009). Mercury isotope fractionation during
628 liquid–vapor evaporation experiments. *Geochim. Cosmochim. Acta* 73: 2693–27113

629

630 Fleming E. J., Mack E. E., Green P. G. and Nelson D. C. (2006). Mercury Methylation from
631 Unexpected Sources: Molybdate-Inhibited Freshwater Sediments and an Iron-Reducing Bacterium.
632 *Appl. Environ. Microbiol.* 72: 457–464

633

634 Fréry N., Maury-Brachet R., Maillot E., Deheeger M., De Merona B. and Boudou A. (2001). Gold-
635 mining activities and mercury contamination of native Amerindian communities in French Guiana:
636 key role of fish in dietary uptake. *Environ. Health Perspect.* 109: 449

637

638 Fu X., Maruszczak N., Wang X., Gheusi F. and Sonke, J. E. (2016). Isotopic composition of gaseous
639 elemental mercury in the free troposphere of the Pic du Midi observatory, France. *Environ. Sci.*
640 *Technol.* 50 (11): 5641–5650

641

642 Gantner N., Hintelmann H., Zheng W. and Muir D. C. (2009). Variations in stable isotope
643 fractionation of Hg in food webs of Arctic lakes. *Environ. Sci. Technol.* 43: 9148–9154

644

645 Gentès S., Coquery M., Vigouroux R., Hanquiez V., Allard L. and Maury-Brachet R. (2019).
646 Application of the European Water Framework Directive: Identification of reference sites and
647 bioindicator fishspecies for mercury in tropical freshwater ecosystems (FrenchGuiana). *Ecological*
648 *Indicators.* 106. 105468

649

650 Gerhke G. E., Blum J. D., Slotton D. G., Greenfield B. K. (2011). Mercury isotopes link mercury in
651 San Francisco Bay forage fish to surface sediments. *Environ. Sci. Technol.* 45: 1264–1270

652

653 Goix S., Maurice L., Laffont L., Rinaldo R., Lagane C., Bosq A., Chmeleff J., Menges J., Heimbürger
654 L. E., Maury-Brachet R. and Sonke J. (2019). A Hg stable isotope study of Hg cycling in the tropical
655 environment and quantification of the gold-mining impact on French Guiana river sediments.
656 *Chemosphere* 219: 684–694.

657

658 Gratz L. E., Keeler G. J., Blum J. D. and Sherman L. S. (2010). Isotopic composition and fractionation
659 of mercury in Great Lakes precipitation and ambient air. *Environ. Sci. Technol.* 44 (20): 7764–7770

660

661 Grimaldi C., Grimaldi M. and Guedron S. (2008). Mercury distribution in tropical soil profiles related
662 to origin of mercury and soil processes. *Sci. Total Environ.* 401: 121–129

663

664 Grimaldi M., Guédron S. and Grimaldi C. (2015). Impact of gold mining on mercury contamination
665 and soil degradation in Amazonian ecosystems of French Guiana. In : Brearley F.Q. (ed.), Thomas

666 A.D. (ed.) Land-use change impacts on soil processes : tropical and savannah ecosystems. Wallingford
667 : CABI, 95-107. ISBN 978-1-78064-210-9
668

669 Guedron S., Grimaldi C., Chauvel C., Spadini L. and Grimaldi M. (2006). Weathering versus
670 atmospheric contributions to mercury concentrations in French Guiana soils. *Appl. Geochem.* 21:
671 2010–2022
672

673 Guedron S., Grangeon S., Lanson B. and Grimaldi M. (2009). Mercury speciation in a tropical soil
674 association; Consequence of gold mining on Hg distribution in French Guiana. *Geoderma* 153: 331–
675 346
676

677 Guimarães J. R. D., Meili M., Malm O. and Maria de Souza Brito E. (1998). Hg methylation in
678 sediments and floating meadows of a tropical lake in the Pantanal floodplain, Brazil. *Sci. Total*
679 *Environ.* 213: 165–175
680

681 Hobson, K. A., Fisk, A., Karnovsky, N., Holst, M., Gagnon, J. M. and Fortier, M. (2002). A stable
682 isotope (d13C, d15N) model for the NorthWater food web: implications for evaluating
683 trophodynamics and the flow of energy and contaminants. *Deep Sea Res. Part II Top. Stud. Oceanogr.*
684 49 (22e23): 5131e5150
685

686 Jiskra, M., Wiederhold, J.G., Skyllberg, U., Kronberg, R.-M., Kretzschmar, R., 2017. Source tracing
687 of natural organic matter bound mercury in boreal forest runoff with mercury stable isotopes. *Environ.*
688 *Sci.: Processes Impacts* 19, 1235–1248.
689

690 Keith, P., Persat, H., Feunteun, E. and Allardi, J. (2011). *Les Poissons d’eau douce de France.*
691 *Collection Inventaires & biodiversité. Biotope Editions, Publications scientifiques du Muséum* 552 (in
692 French)
693

694 Kerin E. J., Gilmour C. C., Roden E., Suzuki M. T., Coates J. D. and Mason R. P. (2006). Mercury
695 Methylation by Dissimilatory Iron-Reducing Bacteria. *Appl. Environ. Microbiol.* 72: 7919–7921
696

697 Klaus, J. E., Hammerschmidt, C. R., Costello, D. M., Burton, G. A. (2016). Net methylmercury
698 production in 2 contrasting stream sediments and associated accumulation and toxicity to periphyton.
699 *Environ. Toxicol. Chem.* 35(7), 1759-1765
700

701 Kwon S. Y., McIntyre P. B., Flecker A. S. and Campbell L. M. (2012a). Mercury biomagnification in
702 the food web of a neotropical stream. *Sci. Total Environ.* 417-418: 92–97
703

704 Kwon S. Y., Blum J. D., Carvan M. J., Basu N., Head J. A., Madenjian C. P. and David S. R. (2012b).
705 Absence of Fractionation of Mercury Isotopes during Trophic Transfer of Methylmercury to
706 Freshwater Fish in Captivity. *Environ. Sci. Technol.* 46: 7527–7534
707

708 Laffont L., Sonke J. E., Maurice L., Hintelmann H., Pouilly M., Sánchez Bacarreza Y., Perez T. and
709 Behra P. (2009). Anomalous mercury isotopic compositions of fish and human hair in the Bolivian
710 Amazon. *Environ. Sci. Technol.* 43: 8985–8990
711

712 Laffont L., Sonke J. E., Maurice L., Monrroy S. L., Chincheros J., Amouroux D. and Behra P. (2011).
713 Hg Speciation and Stable Isotope Signatures in Human Hair As a Tracer for Dietary and Occupational
714 Exposure to Mercury. *Environ. Sci. Technol.* 45: 9910–9916
715

716 Lamberti, G. A. (1996). The role of periphyton in benthic food webs. *Algal ecology*
717

718 Laperche V., Maury-Brachet R., Blanchard F., Dominique Y., Durrieu G., Massabuau J. C., Bouillard
719 H., Joseph B., Laporte P., Mesmer-Dudons N., Duflo V. et Callier L. (2007). Répartition régionale
720 du mercure dans les sédiments et les poissons de six fleuves de Guyane: Rapport BRGM/RP55965-
721 FR, 203.
722

723 Layman C. A., Araujo M. S., Boucek R., Hammerschlag- Peyer C. M., Harrison E., Jud Z. R., Matich
724 P., Rosenblatt A. E., Vaudo J. J., Yeager L. A., Post D. M. and Bearhop S. (2012). Applying stable
725 isotopes to examine food- web structure: an overview of analytical tools. *Biological Reviews* 87 (3):
726 545-562
727

728 Le Bail, P.-Y., Covain, R., Jégu, M., Fisch-Muller, S., Vigouroux R. and Keith, P. (2012). Updated
729 checklist of the freshwater and estuarine fishes of French Guiana. *Cybium* 36 (1): 293–319.
730

731 Le Bail, P. Y., Keith, P. and Planquette, P. (2000). Atlas des poissons d'eau douce de Guyane ; Tome
732 2, fascicule II: Siluriformes. Patrimoines naturels (MNHN/SPN), 43, 307. Paris (in French).
733

734 Lee, C. C. (2005). *Environmental Engineering Dictionary* (Government Institutes)
735

736 Lepak R., Yin R., Krabbenhoft D. P., Ogorek J. M., DeWild J. F., Holsen T. M. and Hurley J. P.
737 (2015). Use of stable isotope signatures to determine mercury sources in the Great Lakes. *Environ.*
738 *Sci. Technol. Lett.* 2 (12): 335–341
739

740 Marshall B. G., Veiga M. M., Kaplan R. J., Adler Miserendino R., Schudel G., Bergquist B. A.,
741 Guimarães J. R. D., Sobral L. G. S. and Gonzalez-Mueller C. (2018). Evidence of transboundary
742 mercury and other pollutants in the Puyango-Tumbes River basin, Ecuador–Peru. *Environ. Sci.:*
743 *Processes Impacts.* 20: 632-641
744

745 Masbou J. (2014). Etude des processus métaboliques, écologiques et biogéochimiques contrôlant le
746 fractionnement isotopique du Hg chez les mammifères marins de l'Arctique. PhD dissertation,
747 Université Toulouse Paul Sabatier, pp 275
748

749 Masbou J., Point D. and Sonke J. E. (2013). Application of a selective extraction method for
750 methylmercury compound specific stable isotope analysis (MeHg-CSIA) in biological materials. *J.*
751 *Anal. At. Spectrom.* 28: 1620
752

753 Mason R. P., Reinfelder J. R. and Morel F. M. M. (1995). Bioaccumulation of mercury and
754 methylmercury. *Water. Air. Soil Pollut.* 80: 915–921
755

756 Maurice L., Goix S., Laffont L., Sonke J. E., Maury-Brachet R., Lagane C., Bosq A., Candaudap F.
757 and Rinaldo R. (2014). Hg stable isotope as a tracer of gold mining activities in the Oyapock river
758 basin, French Guiana. In: 17th ICHMET International Conference on Heavy Metals in the
759 Environment, Sept. 2014, Guiyang, China.

760
761 Maurice-Bourgoin L., Quiroga I., Chincheros J. and Courau P. (2000). Mercury distribution in waters
762 and fishes of the upper Madeira rivers and mercury exposure in riparian Amazonian populations. *Sci.*
763 *Total Environ.* 260: 73-86
764
765 Maurice-Bourgoin L., Quiroga I., Guyot I. L. and Malm O. (1999). Mercury Pollution in the Upper
766 Beni River Basin Bolivia. *Ambio* 28: 302–306
767
768 Maury-Brachet R., Durrieu G., Dominique Y. and Boudou A. (2006). Mercury distribution in fish
769 organs and food regimes: Significant relationships from twelve species collected in French Guiana
770 (Amazonian basin). *Sci. Total Environ.* 368: 262–270
771
772 Maury-Brachet R., Gentes S., Dassié E. P., Feurtet-Mazel A., Vigouroux R., Laperche V., Gonzalez
773 P., Hanquiez V., Mesmer-Dudons N., Durrieu G. and Legeay A. (2020). Mercury contamination levels
774 in the bioindicator piscivorous fish *Hoplias aimara* in French Guiana rivers: mapping for risk
775 assessment. *Environ Sci Pollut Res* 27: 3624–3636
776
777 Meech J. A., Veiga M. M. and Tromans D. (1997). Emission and Stability of Mercury in the Amazon.
778 *Can. Metall. Q.* 36: 231–239
779
780 Mergler, D.; Anderson, H. A.; Chan, L. H. M.; Mahaffey, K. R.; Murray, M.; Sakamoto, M.; Stern, A.
781 H. (2007). Methylmercury exposure and health effects in humans: A worldwide concern. *Ambio* 36:
782 3-11
783
784 Monperrus M., Rodriguez-Gonzalez P., Amouroux D., J. Garcia Alonso I. and Donard O. F. X.
785 (2008). Evaluating the Potential and Limitations of Double-Spiking Species-Specific Isotope Dilution
786 Analysis for the Accurate Quantification of Mercury Species in Different Environmental Matrices.
787 *Analytical and Bioanalytical Chemistry* 390 (2): 655-666
788
789 De Morais L. T. and Raffray J. (1999). Movements of *Hoplias aimara* during the filling phase of the
790 Petit-Saut dam, French Guyana. *J. Fish Biol.* 54: 627–635
791
792 Navarro P., Clémens S., Perrot V., Bolliet V., Tabouret H., Guérin T., Monperrus M. and Amouroux
793 D. (2011). Simultaneous determination of mercury and butyltin species using a multiple species-
794 specific isotope dilution methodology on the European, *Anguilla anguilla* glass eel and yellow eel.
795 *International Journal of Environmental Analytical Chemistry.* 1-17
796
797 Neres- Lima V., Machado- Silva F., Baptista D. F., Oliveira R. B. S., Andrade P. M., Oliveira A. F.,
798 Sasada- Sato C. Y., Silva- Junior E. F., Feijó- Lima R., Angelini R., Camargo P. B. and Moulton T.P.
799 (2017). Allochthonous and autochthonous carbon flows in food webs of tropical forest streams.
800 *Freshwater Biology* 62 (6): 1012-1023
801
802 Obrist D., Agnan Y., Jiskra M., Olson C. L., Colegrove D. P., Hueber J., Moore C. W., Sonke J. E.
803 and Helmig D. (2017). Tundra uptake of atmospheric elemental mercury drives Arctic mercury
804 pollution. *Nature.* 547 (7662): 201–204
805

806 Pacyna E. G., Pacyna J. M., Sundseth K., Munthe J., Kindbom K., Wilson S., Steenhuisen F. and
807 Maxson P. (2010). Global emission of mercury to the atmosphere from anthropogenic sources in 2005
808 and projections to 2020. *Atmos. Environ.* 44: 2487–2499
809

810 PAG, Suivi environnemental des impacts de l'orpaillage illégal. 2017 Bulletin n°5 5p.
811

812 Perrot V., Epov V. N., Pastukhov M. V., Grebenshchikova V. I., Zouiten C., Sonke J. E., Husted S.,
813 Donard O. F. X. and Amouroux D. (2010). Tracing sources and bioaccumulation of mercury in fish of
814 Lake Baikal- Angara River using Hg isotopic composition. *Environ. Sci. Technol.* 44: 8030–8037
815

816 Perrot V., Pastukhov M. V., Epov V. N., Husted S., Donard O. F. X. and Amouroux D. (2012). Higher
817 Mass-Independent Isotope Fractionation of Methylmercury in the Pelagic Food Web of Lake Baikal
818 (Russia). *Environ. Sci. Technol.* 46: 5902–5911
819

820 Peterson, B. J., Fry, B. (1987). Stable isotopes in ecosystem studies. *Annu. Rev. Ecol. Syst.* 18: 293-
821 320
822

823 Pfeiffer W. C. and de Lacerda L. D. (1988). Mercury inputs into the Amazon Region, Brazil. *Environ.*
824 *Technol. Lett.* 9: 325–330
825

826 Planquette P., Keith P. and Le Bail P-Y. (1996). Atlas des poissons d'eau douce de Guyane. Museum
827 d'Histoire Naturelle. Paris
828

829 Post D. M. (2002). Using stable isotopes to estimate trophic position: models, methods, and
830 assumptions. *Ecology* 83: 703–718
831

832 Post, D. M., Layman, C. A., Arrington, D. A., Takimoto, G., Quattrochi, J., Montana, C. G. (2007).
833 Getting to the fat of the matter: models, methods and assumption for dealing with lipids in stable
834 isotope analyses. *Oecologia.* 152: 179-189
835

836 Pouilly M., Rejas D., Perez T., Duprey J-L., Molina C. I., Hubas C. and Guimarães J. R. D. (2013).
837 Trophic structure and mercury biomagnification in tropical fish assemblages, Itenez River, Bolivia.
838 *PLoS ONE* 8(5): e65054
839

840 Rahm M., Thibault P., Shapiro A., Smartt T., Paloeng C., Crabbe S., Farias P., Carvalho R., Joubert P.
841 (2017). Monitoring the impact of gold mining on the forest cover and freshwater in the Guiana Shield.
842 Reference year 2015. REDD+ for the Guiana Shield Project and WWF Guianas. Report pp.20
843

844 Rimbaud D., Restrepo M., Louison A., Boukhari R., Ardillon V., Carles G., Lambert V. and Jolivet A.
845 (2017). Blood lead levels and risk factors for lead exposure among pregnant women in western French
846 Guiana: the role of manioc consumption. *Journal of Toxicology and Environmental Health, Part A*,
847 DOI:10.1080/15287394.2017.1331490
848

849 Roulet M., Lucotte M., Canuel R., Rheault I., Tran S., De Freitas Gog Y.G., Farella N., Souza do Vale
850 R., Sousa Passos C. J., Jesus da Silva E., Mergler D. and Amorim M. (1998). Distribution and
851 partition of mercury in waters of Tapajos. *Sci. Total Environ.* 213: 203–2011
852

853 Roulet M., Lucotte M., Guimarães J. R. D. and Rheault I. (2000). Methylmercury in water, seston, and
854 epiphyton of an Amazonian river and its floodplain, Tapajos River, Brazil. *Sci. Total Environ.* 261:
855 43–59
856
857 Roulet M., Guimaraes J. R. D. and Lucotte M. (2001). Methylmercury production and accumulation
858 in sediments and soils of an Amazonian floodplain—effect of seasonal inundation. *Water. Air. Soil*
859 *Pollut.* 128: 41–60
860
861 Roulet M. and Maury-Brachet R. (2001). Le mercure dans les organismes aquatiques amazoniens. In
862 *Le mercure en Amazonie.* IRD Editions
863
864 Senn D. B., Chesney E. J., Blum J. D., Bank M. S., Maage A. and Shine J. P. (2010). Stable Isotope
865 (N, C, Hg) Study of Methylmercury Sources and Trophic Transfer in the Northern Gulf of Mexico.
866 *Environ. Sci. Technol.* 44: 1630–1637
867
868 Smith R. S., Wiederhold J. G., Jew A. D., Brown G. E., Bourdon B. and Kretzschmar R. (2014).
869 Small-scale studies of roasted ore waste reveal extreme ranges of stable mercury isotope signatures.
870 *Geochim. Cosmochim. Acta* 137: 1–17
871
872 Sonke, J. E., (2011). A global Hg isotope box-model for mass-independent Hg isotope fractionation. .
873 *Geochim. Cosmochim. Acta* 75: 4577-4590
874
875 Sonke, J. E. and Blum, J. D. (2013). Advances in mercury stable isotope biogeochemistry. *Chemical*
876 *Geology* 336: 1-4
877
878 Telmer K., Costa M., Simões Angélica R., Araujo E. S. and Maurice Y. (2006). The source and fate of
879 sediment and mercury in the Tapajós River, Pará, Brazilian Amazon: Ground- and space-based
880 evidence. *J. Environ. Manage.* 81: 101–113
881
882 Tsui M. T. K., Blum J. D., Kwon S. Y., Finlay J. C., Balogh S. J. and Nollet Y. H. (2012). Sources and
883 Transfers of Methylmercury in Adjacent River and Forest Food Webs. *Environ. Sci. Technol.* 46:
884 10957–10964.
885
886 Tsui, M.T.K., Blum, J.D., Finlay, J.C., Balogh, S.J., Kwon, S.Y., Nollet, Y.H., 2013.
887 Photodegradation of methylmercury in stream ecosystems. *Limnol. Oceanogr.* 58, 13–22.
888
889 Tsui M. T. K., Blum J. D. and Kwon S. Y. 2020. Review of stable mercury isotopes in ecology and
890 biogeochemistry. *Science of the Total Environment.* 716: 135386
891
892 US Environmental Protection Agency. (2012). [https://www.epa.gov/fish-tech/epa-fda-fish-advice-](https://www.epa.gov/fish-tech/epa-fda-fish-advice-technical-information)
893 [technical-information](https://www.epa.gov/fish-tech/epa-fda-fish-advice-technical-information)
894
895 Vander Zanden, J. and Rasmussen, J.B. (1996). A trophic position model of pelagic food webs: impact
896 on contaminant bioaccumulation in lake trout. *Ecol. Monogr.* 66 (4): 451e477
897
898 Velásquez-López, P. C. (2010). Mercury in artisanal and small scale gold mining: identifying
899 strategies to reduce environmental contamination in Southern Ecuador. University of British
900 Columbia. doi:10.14288/1.0071256

901
902 WHO. (1990). Environmental health criteria 101.
903 <http://www.inchem.org/documents/ehc/ehc/ehc101.htm>, 02.12.2014
904
905 Woerndle, G.E., Tsui, M.T.K., Sebestyen, S.D., Blum, J.D., Nie, X., Kolka, R.K., 2018. New insights
906 on ecosystem mercury cycling revealed by stable isotopes of mercury in water flowing from a
907 headwater peatland catchment. *Environ. Sci. Technol.* 52, 1854–1861.
908
909 World Wildlife Founds. (2018). Lutte contre l'Orpillage Illégal en Guyane; orientations pour une
910 efficacité renforcée. Analyse WWF France, bureau Guyane.
911 [https://www.wwf.fr/sites/default/files/doc-](https://www.wwf.fr/sites/default/files/doc-201806/180613_Rapport_Lutte_orpillage_illegal_Guyane.pdf)
912 [201806/180613_Rapport_Lutte_orpillage_illegal_Guyane.pdf](https://www.wwf.fr/sites/default/files/doc-201806/180613_Rapport_Lutte_orpillage_illegal_Guyane.pdf)
913
914 Yin R., Feng X., Hurley J. P., Krabbenhoft D. P., Lepak R. F., Kang S., Yang H. and Li X. (2016).
915 Historical records of mercury stable isotopes in sediments of Tibetan Lakes. *Sci. Rep.* 6: 23332