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## Stable mercury concentrations of tropical tuna in the south western Pacific ocean: An 18-year monitoring study

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## 17 **Abstract**

18 Global anthropogenic mercury (Hg) emissions to the atmosphere since industrialization are widely  
19 considered to be responsible for a significant increase in surface ocean Hg concentrations. Still  
20 unclear is how those inputs are converted into toxic methylmercury (MeHg) then transferred and  
21 biomagnified in oceanic food webs. We used a unique long-term and continuous dataset to explore  
22 the temporal Hg trend and variability of three tropical tuna species (yellowfin, bigeye, and skipjack)  
23 from the southwestern Pacific Ocean between 2001 and 2018 (n=590). Temporal trends of muscle  
24 nitrogen ( $\delta^{15}\text{N}$ ) and carbon ( $\delta^{13}\text{C}$ ) stable isotope ratios, amino acid (AA)  $\delta^{15}\text{N}$  values and  
25 oceanographic variables were also investigated to examine the potential influence of trophic,  
26 biogeochemical and physical processes on the temporal variability of tuna Hg concentrations. For the  
27 three species, we detected significant inter-annual variability but no significant long-term trend for  
28 Hg concentrations. Inter-annual variability was related to the variability in tuna sampled lengths  
29 among years and to tuna muscle  $\delta^{15}\text{N}$  and  $\delta^{13}\text{C}$  values. Complementary AA- and model-estimated  
30 phytoplankton  $\delta^{15}\text{N}$  values suggested the influence of baseline processes with enhanced tuna Hg  
31 concentrations observed when dinitrogen fixers prevail, possibly fuelling baseline Hg methylation  
32 and/or MeHg bioavailability at the base of the food web. Our results show that MeHg trends in top  
33 predators do not necessarily capture the increasing Hg concentrations suspected at the global oceanic  
34 scale due to the complex and variable processes governing Hg deposition, methylation,  
35 bioavailability and biomagnification. This illustrates the need for long-term standardized monitoring  
36 programs of marine biota worldwide.

37 **Keywords:** methylmercury, stable isotope data, yellowfin tuna, bigeye tuna, skipjack tuna, New  
38 Caledonia-Fiji region

## 39 **1. Introduction**

40 Mercury (Hg) is a widely distributed trace element of particular concern to human and ecosystem  
41 health. Its toxicological effects are strongly dependent on the physical properties of its different  
42 chemical forms (Hintelmann, 2010). Gaseous elemental Hg is emitted to the atmosphere through  
43 natural (volcanism and erosion) and anthropogenic (fossil fuel combustion and artisanal gold mining)  
44 processes (Pirrone et al., 2010). In open ocean regions, the dominant source of inorganic Hg (iHg) is  
45 atmospheric deposition with other inputs coming from ocean margins, groundwater, benthic  
46 sediments and hydrothermal vents (Selin et al., 2007). Only a fraction of iHg is naturally converted  
47 into methylmercury (MeHg), the organometallic form of Hg characterised by strong neurotoxicity,  
48 persistence and unique biomagnification properties in food webs. Over the past 150 years of the  
49 industrial era, anthropogenic Hg use and emissions have considerably modified the natural global Hg  
50 cycle (Selin et al., 2008). Models suggest that anthropogenic activities have increased atmospheric  
51 Hg concentrations and have tripled the iHg content in the global ocean surface waters (Lamborg et  
52 al., 2014), but with suspected sub-regional differences. In particular in the North Pacific Ocean,  
53 surface water Hg concentrations have been reported to be higher in the eastern North Pacific than in  
54 the western North Pacific (Sunderland et al., 2009). Most global models utilize ocean Hg data  
55 collected in oceanic regions from the northern hemisphere, but questions have risen about how well  
56 these models describe potential hemispherical ocean patterns with lower Hg concentrations reported  
57 in the southern atmosphere (Horowitz et al., 2017).

58 Humans are exposed to MeHg primarily by the consumption of marine fish, especially of top  
59 predators such as tuna (Mergler et al., 2007; Sunderland, 2007) as MeHg biomagnifies naturally in  
60 marine food webs (Cai et al., 2007; Ordiano-Flores et al., 2011; 2012). Mercury concentrations in  
61 tuna are known to vary geographically between ocean basins and species (Chouvelon et al., 2017;  
62 Houssard et al., 2019; Kojadinovic et al., 2006; Nicklisch et al., 2017), sometimes exceeding food  
63 safety guidelines (1 mg.kg<sup>-1</sup> fresh tissue) (WHO and UNEP Chemicals, 2008). Mercury

64 concentrations in organisms are governed by a complex interplay between physiological (age/length,  
65 metabolism and assimilation efficiencies), ecological (foraging depth and food web structures),  
66 biogeochemical (baseline in situ MeHg production and bioavailability) and physical (thermocline  
67 depth and sea surface temperature) processes (Cai et al., 2007; Chouvelon et al., 2017; Choy et al.,  
68 2009; Teffer et al., 2014). Recently, a spatial study from the western Pacific Ocean suggested that  
69 fish length and deeper thermoclines (used as a proxy of tuna foraging habitat) were the two main  
70 drivers enhancing Hg concentrations in tuna, with tuna trophic position (TP) and oceanic primary  
71 production being of less importance but still influencing tuna Hg concentrations (Houssard et al.,  
72 2019).

73 Despite their relatively high MeHg concentrations, tuna species are among the most popular fish  
74 species consumed worldwide (FAO, 2018). In the central and western Pacific Ocean, tuna fisheries  
75 accounted in 2017 for about 80% of the total Pacific Ocean catches and 54% of the global tuna catch  
76 (Williams and Reid, 2018). For Pacific Island countries and territories, the large tuna resources  
77 deliver great economic benefits through the sale of fishing access to distant water fishing nations and  
78 employment in fish processing (Bell et al., 2011; Gillett, 2009). In terms of food and nutrition  
79 security, tuna also represent a major source of proteins, essential fatty acids, vitamins and minerals  
80 (Di Bella et al., 2015; Sirot et al., 2012). These species have been consequently identified as key  
81 food resources for good nutrition to complement declining coastal resources in a context of high  
82 levels of diabetes and obesity in this region, while taking into account their Hg content (Bell et al.,  
83 2015).

84 Anticipating changes in human Hg exposure depends on our ability to capture and predict spatial and  
85 temporal Hg trends in marine food webs. Only two temporal studies of tuna Hg content are available  
86 to date, showing distinct results regarding Hg long-term trends. In the northern central Pacific Ocean  
87 (Hawaii), Drevnick et al. (2015) suggested that Hg concentrations in yellowfin tuna (*Thunnus*  
88 *albacares*) increased by at least 3.8% per year between 1971 and 2008, despite considering only

89 three sampling years. Conversely, in the North Atlantic Ocean, Lee et al. (2016) revealed a decadal  
90 decline of Hg concentrations in Atlantic bluefin tuna (*T. thynnus*) of about 2.4 % between 2004 and  
91 2012, suggesting potential benefits of the reduction of anthropogenic emission in North America.  
92 Unfortunately, no complementary trophic ecology data were available in those two studies to discuss  
93 if confounding ecological factors might contribute to the contrasted temporal trends of Hg content  
94 documented worldwide.

95 Stable isotope ratios of nitrogen ( $\delta^{15}\text{N}$ ) and carbon ( $\delta^{13}\text{C}$ ) are widely used to examine trophic  
96 ecology of marine organisms (Fry, 2006). In particular,  $\delta^{13}\text{C}$  values provide information on basal  
97 organic carbon sources while  $\delta^{15}\text{N}$  values are used to estimate the TP of a consumer as they increase  
98 predictably between prey and consumers. Therefore,  $\delta^{15}\text{N}$  values are commonly used to explore Hg  
99 biomagnification along trophic webs (Atwell et al., 1998; Cai et al., 2007; Teffer et al., 2014). In  
100 addition to diet (i.e. trophic effects), variability of basal stable isotopic composition (i.e. baseline  
101 effects) also affects consumer  $\delta^{15}\text{N}$  values (Lorrain et al., 2015b). Thus,  $\delta^{15}\text{N}$  values also represent a  
102 potential proxy to infer baseline biological processes fuelling Hg methylation and/or net basal MeHg  
103 bioavailability. To account for complex primary production dynamics influencing the isotopic  
104 baseline, and produce accurate measures of a consumer's TP, amino acid compound-specific  $\delta^{15}\text{N}$   
105 analyses (AA-CSIA) are also used (Choy et al., 2015; Lorrain et al., 2015b). Within a consumer, the  
106  $\delta^{15}\text{N}$  values of source amino acids (Sr-AA; e.g., phenylalanine and glycine) track the  $\delta^{15}\text{N}$  values at  
107 the base of the food web, while trophic AA (Tr-AA; e.g., glutamic acid), being enriched in  $^{15}\text{N}$  with  
108 each trophic level, provide information about a consumer's TP (Popp et al., 2007). Overall, the  
109 combination of muscle stable isotope ratios and AA-CSIA is a powerful tool to investigate both Hg  
110 methylation and biomagnification along food webs.

111 The specific objectives of our study were to i) investigate long-term trends of Hg concentrations in  
112 tuna from the New Caledonia-Fiji sub-region in the southwestern Pacific Ocean, and to ii) identify  
113 the main drivers of inter-annual variability of tuna Hg concentrations among physiological,

114 ecological, biogeochemical and physical parameters. We analysed a unique long-term, continuous  
115 and large Hg dataset (n=590) in three commercial tropical tuna species (yellowfin, *Thunnus*  
116 *albacares*; bigeye, *T. obesus*; and skipjack, *Katsuwonus pelamis*) from 2001 to 2018.  
117 Complementary muscle  $\delta^{15}\text{N}$  and  $\delta^{13}\text{C}$  values, AA-CSIA and model estimated phytoplankton  $\delta^{15}\text{N}$   
118 values were also used to explore the possible influence of biogeochemical and ecological factors on  
119 Hg content in tuna (i.e. changes in nutrient sources, primary productivity and tuna TP reflecting  
120 pathways of energy transfer). Finally, a suite of oceanographic variables (sea surface temperature,  
121 chlorophyll-a, net primary production, mixed layer depth, depths of isotherms 20°C and 12°C and  
122 oceanic El Niño index) were also included as potential explanatory factors of inter-annual variability  
123 in tuna Hg concentrations.

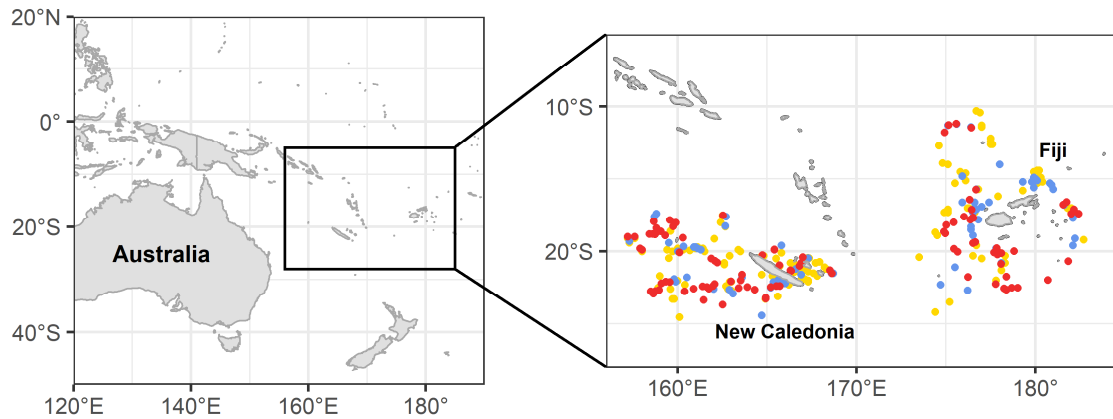
## 124 **2. Materials and Methods**

### 125 **2.1. Study area**

126 Within the large oceanic region of the south western Pacific Ocean, the New Caledonia-Fiji sub-  
127 region is characterized by similar marine systems in terms of ocean dynamics, phytoplankton,  
128 zooplankton, and micronekton (Le Borgne et al., 2011). Oceanographic parameters are known to be  
129 driven by wind regimes, in particular southeast oriented trade winds and northwest oriented winds,  
130 associated with the monsoon season. During austral summers (from December to May), the trade  
131 winds become prevalent while the north-west oriented winds are more important during austral  
132 winters (from May to December) (Cravatte et al., 2015; Lorrain et al., 2015a). Compared to other  
133 regions in the Pacific Ocean where primary production mainly relies on  $\text{NO}_3^-$  (with particulate  
134 organic matter (POM)  $\delta^{15}\text{N}$  values  $\sim 4\text{‰}$ ), our study area is known to be a hotspot of  $\text{N}_2$  fixation  
135 (POM  $\delta^{15}\text{N}$  values  $\sim 1\text{‰}$ ), with diazotrophy showing some spatial and large seasonal variability  
136 (Bonnet et al., 2017; Garcia et al., 2007; Shiozaki et al., 2014). In the sub-region of New Caledonia-  
137 Fiji, a significant and unique effort of collecting and archiving tuna samples has been conducted  
138 since 2001 within the Pacific Marine Specimen Bank  
139 (<https://www.spc.int/ofp/PacificSpecimenBank>). Previous analyses of specimens from this tissue  
140 bank showed homogeneous Hg values in the New Caledonia and Fiji region (Houssard et al., 2019),  
141 which justifies the consideration of this sub-region to address temporal trends. Furthermore,  
142 Houssard et al. (2017) showed that tuna were relatively resident at this sub-regional scale as  
143 demonstrated by the similar isotope patterns between particulate organic matter and tuna in New  
144 Caledonia and Fiji.

145





146

147 Figure 1: Sample location of bigeye (blue), yellowfin (yellow), and skipjack (red) caught around New Caledonia and Fiji.

148

## 149 2.2. Sample and data collection

150 **2.2.1. Tuna sampling:** Tuna samples were taken from the Pacific Marine Specimen Bank,  
 151 corresponding to 326 yellowfin, 116 bigeye and 148 skipjack tuna, spanning the 2001-2018 time  
 152 period ( $n_{\text{total}}=590$ ) (Table S1). Sampling was performed onboard commercial fishing boats (longline)  
 153 by trained scientific observers from the National Observer Programs of the Pacific Island Countries  
 154 and Territories. Specimens were selected from 10°S to 25°S and from 157°E to 176°W, covering the  
 155 Economic Exclusive Zone (EEZ) of both New Caledonia and Fiji (Fig. 1). Fork length (FL) was  
 156 measured to the lowest cm and ranged respectively for yellowfin, bigeye and skipjack from 60 to 160  
 157 cm ( $121 \pm 18$  cm; mean  $\pm$  SD), 64 to 160 cm ( $108 \pm 21$  cm; mean  $\pm$  SD) and 42 to 90 cm ( $72 \pm 7$  cm;  
 158 mean  $\pm$  SD). For each fish, a white muscle sample was collected from the anal area and stored frozen  
 159 at -20°C prior to analyses.

160

161 **2.2.2. Environmental variables:** Seven oceanographic variables shown to influence variability of  
 162 Hg concentrations in tropical tuna (Houssard et al., 2019) were used in this study to explore the  
 163 physical drivers of Hg concentrations at the surface and at depth. Surface variables included monthly

164 mean sea surface temperature (SST in °C) from the National Oceanic and Atmospheric  
165 Administration (NOAA, <https://www.ncdc.noaa.gov/oisst>; (Reynolds et al., 2002) and monthly  
166 chlorophyll-a observations (Chl-a, mg.m<sup>-3</sup>) from a continuous dataset of merged L4 Ocean Colour  
167 products provided by GlobColour (<http://globcolour.info>). Monthly means of net primary production  
168 (NPP, mg C.m<sup>-2</sup>.day<sup>-1</sup>) were derived from a vertically generalized production model (VGPM,  
169 <http://www.science.oregonstate.edu/ocean.productivity/custom.php>; Behrenfeld and Falkowski,  
170 1997). Variables at depth or sub-surface included the mixed layer depth (MLD) and depths of the  
171 12°C and 20°C isotherms (D<sub>iso12</sub> and D<sub>iso20</sub>, m), all obtained from the monthly global ARMOR3D L4  
172 dataset (Guinehut et al., 2012). The Oceanic Niño Index (ONI), used to monitor the El Niño-  
173 Southern Oscillation (ENSO), was also collected over the study period. Except for this last variable,  
174 all monthly data were extracted on a 1° x 1° grid from 2001 to 2018. Assuming the relative oceanic  
175 homogeneity of the New Caledonia-Fiji region (Houssard et al., 2017; Le Borgne et al., 2011),  
176 physical variables were averaged over the entire study area to examine only temporal relationships  
177 between tuna Hg concentrations and the environment. Furthermore, given the long process of MeHg  
178 bioaccumulation in tuna muscle (Kwon et al., 2016), we assumed that Hg values of a given  
179 individual captured at a single date and place is not exclusively explained by the environmental  
180 conditions at this date, but also by the conditions prevailing during the previous months or years. As  
181 <sup>15</sup>N turnover (~six months) in tuna white muscle is known to be shorter than MeHg turnover  
182 (Madigan et al., 2012), all oceanographic variables were averaged over a six-month period preceding  
183 individual capture date to explain both Hg and nitrogen isotope data.

184

185 **2.2.3. Estimates of baseline phytoplankton δ<sup>15</sup>N values:** To explore the potential relationship  
186 between the nitrogen cycle and change in tuna Hg concentrations at the top of the food web, baseline  
187 phytoplankton δ<sup>15</sup>N values were estimated from a model of ocean biogeochemistry and isotopes  
188 (MOBI, Somes et al., 2017) at each tuna sample location and year from a hindcast simulation. The

189 biogeochemical component is a 2N2PZD (2 Nutrients, 2 Phytoplankton, 1 Zooplankton, and 1  
190 Detritus) ecosystem model (Somes and Oschlies, 2015). The processes in the model that fractionate  
191 the nitrogen isotopes (i.e. preferentially incorporate  $^{14}\text{N}$  into the product) are phytoplankton  $\text{NO}_3$   
192 assimilation (6 ‰), zooplankton excretion (4 ‰),  $\text{N}_2$  fixation (1 ‰), water column denitrification  
193 (20 ‰) and benthic denitrification (6 ‰), in which the respective fractionation factor yields the  $\delta^{15}\text{N}$   
194 difference between substrate and product (Somes et al., 2010). The baseline  $\delta^{15}\text{N}$  values reproduce  
195 the major features in a global seafloor  $\delta^{15}\text{N}$  database (Tesdal et al., 2013).

196

## 197 **2.3. Analytical methods**

198 **2.3.1. Total mercury concentration analysis:** Total Hg concentrations were analysed on 590  
199 samples: 326 yellowfin, 116 bigeye and 148 skipjack. The majority (n=458) of total Hg  
200 concentrations were performed on homogenized freeze-dried samples by thermal decomposition,  
201 gold amalgamation and atomic adsorption detection (DMA-80, Milestone, Italy) at GET (Toulouse,  
202 France). Blanks and two biological standard reference materials, TORT-3 (lobster hepatopancreas;  
203  $\text{Hg} = 292 \pm 22 \text{ ng.g}^{-1} \text{ dw}$ ) and IAEA-436 (tuna fish flesh homogenate;  $\text{Hg} = 4190 \pm 360 \text{ ng.g}^{-1} \text{ dw}$ ),  
204 covering a wide range of Hg concentrations, were routinely used in each analytical batch to check  
205 Hg measurement accuracy. Complementary Hg data (n=132, only skipjack tuna) were analysed at  
206 the IRD laboratory in Noumea (New Caledonia) by hot plate acidic digestion ( $\text{HNO}_3\text{-H}_2\text{O}_2$ ) followed  
207 by Cold Vapor Atomic Fluorescence Spectroscopy. Blanks and one biological standard reference  
208 materials, DOMR-4 (fish protein;  $\text{Hg} = 412 \pm 36 \text{ ng.g}^{-1} \text{ dw}$ ) were routinely used in each analytical  
209 batch to check Hg measurement accuracy and traceability. Hg contents are expressed on a dry weight  
210 basis (dw).

211

212 **2.3.2. Bulk muscle and compound-specific stable isotope analysis:** Muscle stable isotope ratios  
213 were measured on the 590 samples analysed in Hg but also on complementary samples available  
214 from our study area (n=85) but in too limited quantities to perform both Hg and stable isotope  
215 analyses. In total, 675 samples were therefore analysed for stable isotopes: 360 yellowfin, 150 bigeye  
216 and 165 skipjack.  $\delta^{15}\text{N}$  and  $\delta^{13}\text{C}$  values were obtained from ~1 mg homogenized freeze-dried  
217 samples packed in tin cups and were analyzed using a Costech elemental analyser coupled to an  
218 isotope ratio mass spectrometer (Thermo Scientific Delta Advantage with a Conflo IV interface) at  
219 Union College (New York, USA). Reference standards (EA Consumables sorghum flour ( $\delta^{13}\text{C} = -$   
220  $13.78 \pm 0.17$ ,  $\delta^{15}\text{N} = 1.58 \pm 0.15$ ), in house acetanilide ( $\delta^{13}\text{C} = -34.07$ ,  $\delta^{15}\text{N} = -0.96$ ), IAEA-N-2  
221 ammonium sulfate ( $\delta^{15}\text{N} = 20.3 \pm 0.2$ ), and IAEA-600 caffeine ( $\delta^{13}\text{C} = -27.771 \pm 0.043$ ,  $\delta^{15}\text{N} = 1.0 \pm$   
222  $0.2$ )) were used for isotopic corrections, and to assign the data to the appropriate isotopic scale with  
223 analytical precision better than 0.1‰. Percent C and N were calculated using additional acetanilide  
224 standards of varying mass. Corrections were done using a regression method. Results were reported  
225 in the  $\delta$  unit notation and expressed as parts per thousand (‰) relative to international standards  
226 (atmospheric  $\text{N}_2$  for nitrogen and Vienna Pee Dee belemnite (VPDB) for carbon). Reproducibility of  
227 several samples measured in triplicate was <0.2 ‰. For samples with elevated lipid content (C:N >  
228 3.5),  $\delta^{13}\text{C}$  values were corrected using a mass balance equation with parameters derived from  
229 Atlantic bluefin tuna muscle (Logan et al., 2008).

230 For AA-CSIA, we used both published (Houssard et al., 2017) and newly-analysed amino acid  
231 compound-specific  $\delta^{15}\text{N}$  data on yellowfin tuna (n=10) which had the most robust time series among  
232 the three tuna species on individuals collected in 2003, 2007, 2010, 2013, 2016 and 2018 (n=16). To  
233 do so, the freeze-dried samples were prepared by acid hydrolysis followed by esterification and  
234 trifluoroacetylation as per Dale et al. (2011). The  $\delta^{15}\text{N}$  isotope values of individual amino acids were  
235 determined with a Trace GC gas chromatograph interfaced with a Delta V Plus isotope ratio mass  
236 spectrometer (IRMS) through a GC-C combustion furnace (980°C), reduction furnace (650°C) and

237 liquid N<sub>2</sub> cold trap. The samples (0.5 µl) were injected splitless onto a forte BPX5 capillary column  
 238 (30 m × 0.32 mm × 1.0 µm film thickness) at an injector temperature of 180°C with a constant  
 239 helium flow rate of 1.5 ml min<sup>-1</sup>. The column was initially held at 50°C for 2 min and then increased  
 240 to 120°C at a rate of 10°C.min<sup>-1</sup>. Once at 120°C, the temperature was increased at a rate of 4°C.min<sup>-1</sup>  
 241 to 195°C and then at 5°C.min<sup>-1</sup> to 235°C where it was held for 5 min. The temperature was then  
 242 further increased to 300°C at 15°C.min<sup>-1</sup> and held for 8 min. All samples were analysed at least in  
 243 triplicate. The δ<sup>15</sup>N values were normalised as follows: each sample analysis consisted of three  
 244 separate IRMS analyses bracketed by a suite of amino acids with known δ<sup>15</sup>N values. The slope and  
 245 intercept of known versus measured values were then used to correct the measured values for the  
 246 sample set. Reproducibility associated with isotopic analysis of glutamic acid and phenylalanine  
 247 averaged ± 0.44 ‰ (1 SD) and ranged from ± 0.06 ‰ to ± 0.85 ‰ respectively.

248

249 **2.3.3. Trophic position estimates:** Tuna TP was estimated using the difference of δ<sup>15</sup>N values in  
 250 trophic (Tr-AA) and source (Sr-AA) amino acids, obtained by the following equation:

$$251 \quad TP_{Tr-Sr} = \frac{\delta^{15}N_{Tr-AA} - \delta^{15}N_{Sr-AA} + \beta_{Tr-Sr}}{TEF_{Tr-Sr}} + 1$$

252

253 where δ<sup>15</sup>N<sub>Sr-AA</sub> is the weighted average of glycine and phenylalanine δ<sup>15</sup>N<sub>AA</sub> values and  
 254 δ<sup>15</sup>N<sub>Tr-AA</sub> the weighted average of alanine, glutamic acid, leucine and proline δ<sup>15</sup>N<sub>AA</sub> values.  
 255 β<sub>Tr-Sr</sub> is the difference between Tr-AA and Sr-AA in primary producers and TEF<sub>Tr-Sr</sub> is the <sup>15</sup>N  
 256 enrichment between Tr-AA and Sr-AA per TP. β<sub>Tr-Sr</sub> and TEF<sub>Tr-Sr</sub> were set respectively at 3.6 and  
 257 5.7 (Bradley et al., 2015). Using the weighted average of Sr-AA and Tr-AA reduced uncertainty due  
 258 to the possible large variations of δ<sup>15</sup>N<sub>AA</sub> values (Hayes et al., 1990). Uncertainty in TP estimates  
 259 was calculated by propagation of errors according to Bradley et al. (2015) with a mean error of 0.4.

## 260        **2.4. Statistical analysis**

261    Distribution differences of Hg concentrations, muscle  $\delta^{15}\text{N}$  and  $\delta^{13}\text{C}$  values between species were  
262    first tested with a Kruskal-Wallis test followed by Dunn's post hoc test as hypothesis of normality  
263    was not met.

264

265    **2.4.1. Transformation of mercury concentrations:** Tuna Hg concentrations were log-transformed  
266    to guarantee the homogeneity of variance (Zuur et al., 2010). Further, as Hg is known to  
267    bioaccumulate with length (and age), a power-law relationship ( $\log(\text{Hg}) = a \times (\text{FL} - b)^c - d$ ) was  
268    fit between  $\log(\text{Hg})$  and fish length (fork length, FL) to characterize the bioaccumulative processes  
269    in each tuna species and remove this length effect in further analysis. This allowed the influence of  
270    other potential factors governing Hg concentrations in tuna to be investigated. Residuals from the  
271    length-based Hg model (i.e. observed values – predicted values) were extracted and used to calculate  
272    length-standardized Hg concentrations (at mean species lengths, i.e. FL=100 cm for yellowfin and  
273    bigeye and at FL=70 cm for skipjack), thereafter defined as “standardized Hg concentrations”.

274

275    **2.4.2. Temporal trend and structure analysis:** Temporal patterns of Hg concentrations in tuna  
276    were examined with Moran's eigenvectors maps (MEM) which are derived from a spectral  
277    decomposition of the temporal relationships among the sampling dates (Dray et al., 2006). This  
278    decomposition generates orthogonal eigenfunctions that can then be used in statistical models as  
279    explanatory variables representing the temporal pattern observed. Species-specific temporal  
280    eigenfunction analyses were conducted on  $\log(\text{Hg})$  and standardized Hg concentrations separately  
281    following the method of Legendre and Gauthier (2014). To account for the seasonality in our study  
282    region, each variable was aggregated by year and season, resulting in 36 season/year couples (i.e.  
283    austral winter and austral summer per year from 2001 to 2018). Briefly, long-term trends of Hg

284 concentrations were first tested on seasonal means of Hg concentrations with a redundancy analysis  
285 (RDA) followed by an ANOVA like permutation test. Secondly, to investigate the temporal structure  
286 of log(Hg) and standardized Hg concentrations, we built distance-based matrices among the  
287 sampling periods and distance-based MEMs (dbMEM) eigenfunctions. As sampling design was not  
288 regular for the three tuna species, dbMEMs were built over the 36 seasons, then the seasons with no  
289 sample for a species were removed as recommended with irregularly designed sampling surveys  
290 (Brind'Amour et al., 2018). dbMEMs modeling positive (i.e. observations that are closer in time tend  
291 to display values that are more similar than observations paired at random) or negative temporal  
292 correlation between seasons were selected and tested with redundancy analysis (RDA) followed by  
293 ANOVA like permutation tests to reveal any temporal structure in the response variables. Temporal  
294 structure of Hg concentrations was investigated only for yellowfin and skipjack as there was not  
295 enough data per season for bigeye.

296 For potential ecological and environmental drivers of Hg concentrations, only long-term trends were  
297 investigated as no particular seasonal structure was revealed for both log(Hg) and standardized Hg  
298 concentrations. RDA followed by ANOVA like permutation tests were then fitted separately on tuna  
299 muscle  $\delta^{15}\text{N}$  and  $\delta^{13}\text{C}$  values, and on each environmental variable (SST, MLD, Chl-a, NPP,  $D_{\text{iso}12}$   
300 and  $D_{\text{iso}20}$ ). For TP estimates and Sr-AA  $\delta^{15}\text{N}$  values, as few data were available, long-term trends  
301 were investigated using a linear regression fitted along years.

302

### 303 **2.4.3. Effects of physical and ecological drivers on the temporal variability of Hg content:**

304 Generalized additive models (GAM) were used to test the effects of potential predictors on temporal  
305 variations of Hg concentrations following the formulae:

$$306 \quad Y = \alpha + s_1(X_1) + s_2(X_2) + \dots + s_n(X_n) + \varepsilon$$

307 where  $Y$  is the expected value of the response variable (i.e.  $\log(\text{Hg})$  or standardized Hg  
308 concentrations),  $\alpha$  is the model intercept,  $s_i(X_i)$  is a thin-plate-spline smooth function of the  
309 explanatory variable  $i$ , and  $\varepsilon$  is the error term. Standardized Hg concentrations were assumed to  
310 follow a Gamma distribution while  $\log(\text{Hg})$  a Gaussian one. Explanatory variables tested included  
311 surface (SST, Chl-a, NPP and ONI) and deep ( $D_{\text{iso12}}$ ,  $D_{\text{iso20}}$  and MLD) oceanographic variables as  
312 well as ecological ( $\delta^{15}\text{N}$  and  $\delta^{13}\text{C}$  values) factors. Fish length was also added in the models testing  
313  $\log(\text{Hg})$ . Before performing model computation, variance inflation factors (VIF) were calculated  
314 between all explanatory variables to detect collinearity. Covariates with the highest VIF were  
315 subsequently removed until the highest VIF value was  $< 5$  (Zuur et al., 2010). With this method,  
316 MLD and Chl-a were found to be collinear to other variables and were then removed from the  
317 explanatory variables.  $D_{\text{iso12}}$  and  $D_{\text{iso20}}$  were highly correlated, so only separate models using either  
318  $D_{\text{iso12}}$  or  $D_{\text{iso20}}$  were tested. Explanatory variables were fitted in the GAM with a low spline  
319 complexity ( $k=3$ ) to reduce over-fitting. A backward selection approach was used and we chose the  
320 model with the lowest Akaike's Information Criterion corrected for small samples sizes (AICc,  
321 Burnham and Anderson, 2004). Finally, for each best-fit GAM, assumptions of residuals temporal  
322 trend and auto-correlation were examined graphically with diagnostic plots. The deviance explained  
323 (% DE) for each model was compared to assess predictive capacity. To determine the amount of  
324 variation explained by each explanatory variable, we fitted a separate model for individual variable.  
325 GAM were fitted in R using the mgcv package (Wood and Wood, 2015).

326

327 **2.4.4. Effects of baseline processes on tuna Hg concentrations and tuna  $\delta^{15}\text{N}$  values:** To  
328 investigate the potential influence of baseline processes, in particular the effect of different nitrogen  
329 sources ( $\text{NO}_3^-$ ,  $\text{NO}_2$ ) fuelling primary productivity in this region (Bonnet et al., 2017; Garcia et al.,  
330 2007; Shiozaki et al., 2014), we fitted linear regressions between MOBI estimates of phytoplankton  
331  $\delta^{15}\text{N}$  values and both standardized Hg concentrations and muscle  $\delta^{15}\text{N}$  values in the three tuna



332 species. Complementary linear regressions were fitted on yellowfin samples analysed in  $\delta^{15}\text{N}$  AA-  
333 CSIA to compare standardized Hg concentrations, tuna muscle  $\delta^{15}\text{N}$  values and phytoplankton  $\delta^{15}\text{N}$   
334 estimates regarding Sr-AA  $\delta^{15}\text{N}$  values, used as proxies of baseline nitrogen isotope values. All  
335 statistical analyses were performed with R 3.6.1 (R Core Team, 2018).

### 336 **3. Results**

#### 337 **3.1. Patterns and temporal variability of mercury concentrations**

338 Mercury concentrations (mean  $\pm$  SD, min-max, dw) differed according to species (Kruskal-Wallis,  
339  $p < 0.001$ ) with significantly higher levels in bigeye ( $2.7 \pm 1.7 \mu\text{g}\cdot\text{g}^{-1}$ , 0.3-8.6  $\mu\text{g}\cdot\text{g}^{-1}$ ; Dunn's test,  
340  $p < 0.001$ ) than in yellowfin ( $0.7 \pm 0.5 \mu\text{g}\cdot\text{g}^{-1}$ , 0.1-5.1  $\mu\text{g}\cdot\text{g}^{-1}$ ) and in skipjack ( $0.7 \pm 0.3 \mu\text{g}\cdot\text{g}^{-1}$ , 0.2-1.7  
341  $\mu\text{g}\cdot\text{g}^{-1}$ ) (Figs. 2a & S1). Fish length and  $\log(\text{Hg})$  were positively correlated and fish length explained  
342 respectively 45%, 39% and 18% of the muscle Hg variations in bigeye, yellowfin and skipjack.  
343 Coefficients (a, b, c and d) of the power-law relationships are specified per species in Figure 2c.

344 Inter-annual variability from 2001 to 2018 was detected in both log-transformed and standardized Hg  
345 concentrations for each tuna species, yet no significant long-term temporal trends were detected  
346 (Figs. 2a & 2d; Table 1). Furthermore, models based upon the MEM were not significant for any  
347 variable and any tuna species (Table 1). This illustrates the lack of any significant seasonal structure  
348 of  $\log(\text{Hg})$  and standardized Hg concentrations between 2001 and 2018 in the south western Pacific  
349 Ocean.

350

#### 351 **3.2. Temporal variability of tuna isotopic ratios and trophic position**

352 Muscle  $\delta^{15}\text{N}$  values varied between the three tuna species (Kruskal-Wallis,  $p < 0.001$ ; Dunn's test,  
353  $p < 0.001$  between all pairs of species). Highest  $\delta^{15}\text{N}$  values were found in bigeye ( $12.6 \pm 1.6 \text{‰}$ ),  
354 intermediate values in yellowfin ( $10.8 \pm 1.8 \text{‰}$ ) and lowest values in skipjack ( $9.9 \pm 1.4 \text{‰}$ ) (Figs. 2e  
355 & S1). For the three species, inter-annual variability between 2001 and 2018 was detected on muscle  
356  $\delta^{15}\text{N}$  values but no increasing or decreasing long-term trends were found (Fig. 2e; Table 1).

357 Similarly,  $\delta^{13}\text{C}$  values differed between species (Kruskal-Wallis,  $p < 0.001$ ; Dunn's test,  $p < 0.01$   
358 between all pairs of species) with highest values in bigeye ( $-16.2 \pm 1.0 \text{‰}$ ), intermediate values in

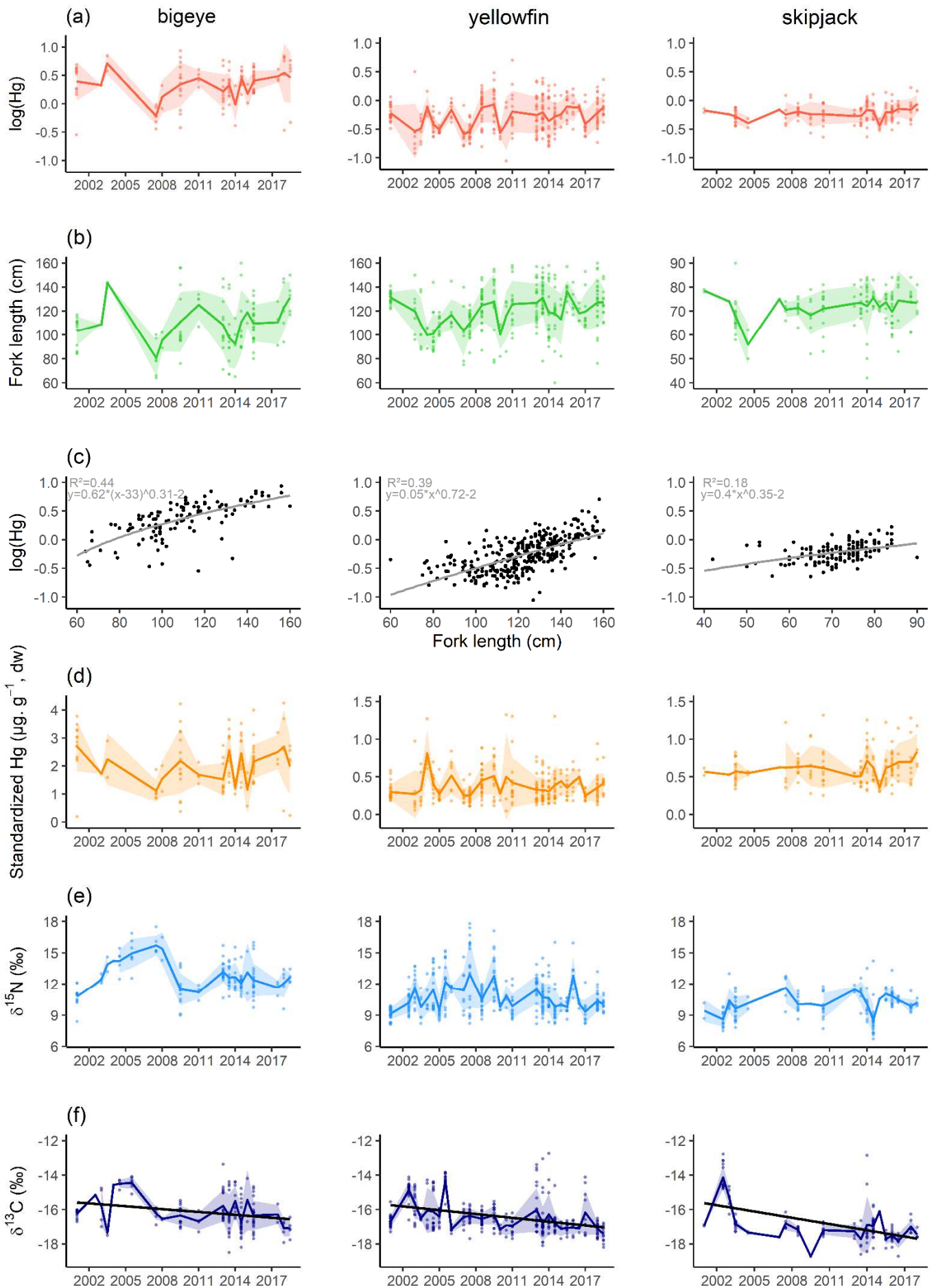
359 yellowfin ( $-16.4 \pm 1.0 \text{ ‰}$ ) and lowest values in skipjack ( $-16.8 \pm 1.1 \text{ ‰}$ ) (Figs 2f & S1). Contrary to  
 360 muscle  $\delta^{15}\text{N}$  values,  $\delta^{13}\text{C}$  values were found to decrease significantly between 2001 and 2018 by a  
 361 mean annual rate of  $0.08 \text{ ‰}$  for bigeye and skipjack and of  $0.07 \text{ ‰}$  for yellowfin (Fig. 2f; Table 1).  
 362 TP estimates and Sr-AA  $\delta^{15}\text{N}$  values in yellowfin varied respectively from 3.4 to 5.5 ( $4.5 \pm 0.5$ ) and -  
 363 5.1 to 11.7  $\text{‰}$  ( $0.4 \pm 3.9$ ) (Fig. 3). Like muscle  $\delta^{15}\text{N}$  values, they varied inter-annually between 2003  
 364 and 2018, however they showed no significant long-term trend ( $p > 0.05$ ).

365

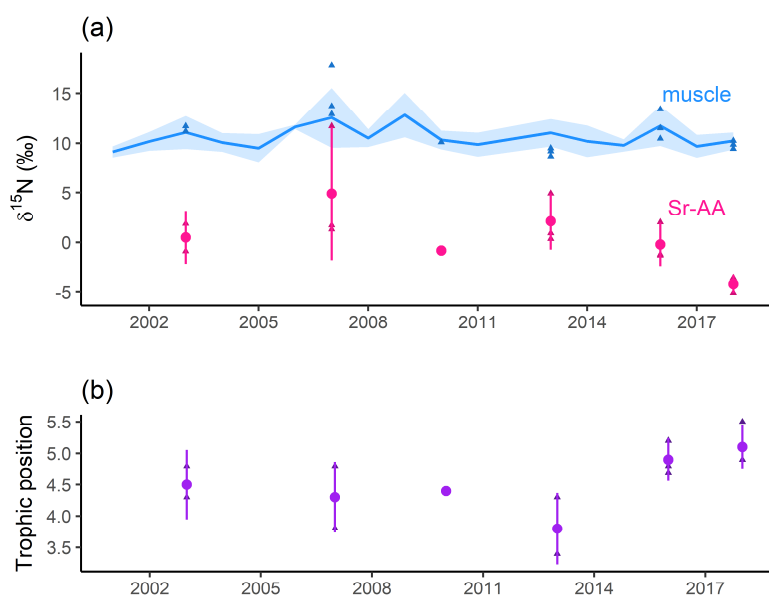
366 Table 1: ANOVA like permutation results to test temporal trend and structure of log(Hg), standardized Hg  
 367 concentrations, muscle  $\delta^{15}\text{N}$  values and  $\delta^{13}\text{C}$  values of tropical tuna. FL: fork length; n: number of tuna individuals; *F*  
 368 and *p*-value: statistics of the tests. \* indicates significant temporal trend.

Species	Response variable	FL (cm) min-max	n	Temporal trend		MEM modelling positive correlation		MEM modelling negative correlation	
				<i>F</i>	<i>p</i> -value	<i>F</i>	<i>p</i> -value	<i>F</i>	<i>p</i> -value
bigeye	log(Hg)		116	0.088	0.800	----- Not enough data -----			
	standardized Hg	64-160	116	0.055	0.799	----- Not enough data -----			
	$\delta^{15}\text{N}$		163	0.132	0.254	----- Not tested -----			
	$\delta^{13}\text{C}$		163	4.043	0.044 *	----- Not tested -----			
yellowfin	log(Hg)		326	4.340	0.056	0.678	0.760	2.495	0.131
	standardized Hg	60-160	326	0.026	0.888	0.4903	0.887	2.15	0.202
	$\delta^{15}\text{N}$		386	0.019	0.580	----- Not tested -----			
	$\delta^{13}\text{C}$		386	10.651	0.005 *	----- Not tested -----			
skipjack	log(Hg)		148	1.470	0.247	666.170	0.053	--- Not enough data ---	
	standardized Hg	42-90	148	2.248	0.169	54.523	0.127	--- Not enough data ---	
	$\delta^{15}\text{N}$		165	1.186	0.308	----- Not tested -----			
	$\delta^{13}\text{C}$		165	4.321	0.041 *	----- Not tested -----			

369



371 Figure 2: Time series of a) log(Hg), b) tuna fork length (cm), d) standardized Hg concentrations ( $\mu\text{g}\cdot\text{g}^{-1}$ , dw), e) tuna  
 372 muscle  $\delta^{15}\text{N}$  values (‰) and f) tuna muscle  $\delta^{13}\text{C}$  values (‰) (the black lines represent the significant temporal trends). c)  
 373 power-law relationships between log(Hg) and fork length;. The coloured lines and shadows give respectively the  
 374 seasonal means and standard deviations. The dots are the observation values.  
 375

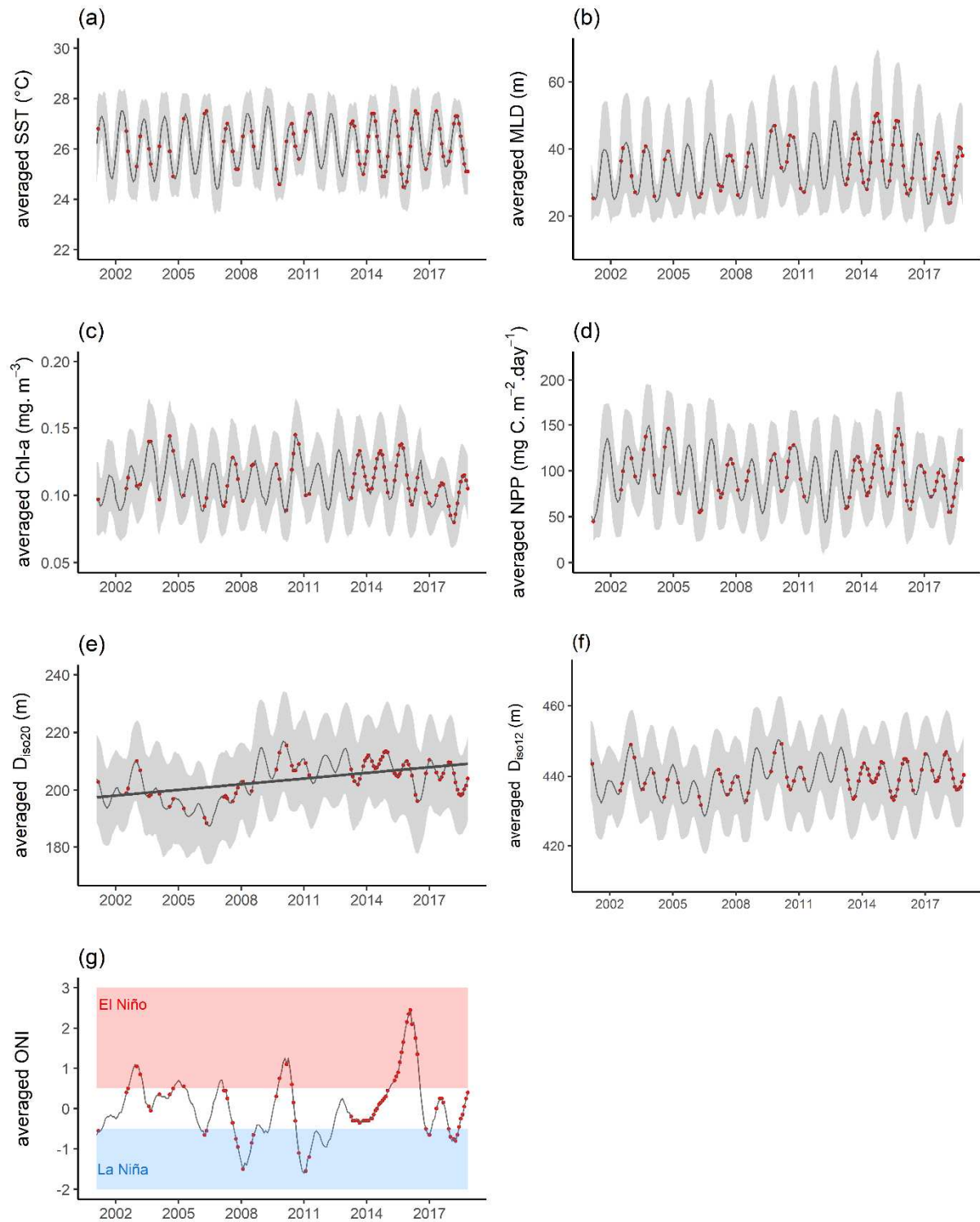


376  
 377 Figure 3: a) Time series of muscle  $\delta^{15}\text{N}$  values (blue), source amino acid (Sr-AA)  $\delta^{15}\text{N}$  values (pink) (‰) and b) trophic  
 378 position (TP) estimates (purple) in yellowfin. The blue line and the blue shadow give respectively the annual means and  
 379 standard deviation of muscle  $\delta^{15}\text{N}$  values. The dots and lines represent respectively the mean values and the standard  
 380 deviations of Sr-AA  $\delta^{15}\text{N}$  values and TP estimates. The triangles are the observations for the selected yellowfin samples  
 381 analysed for AA-CSIA.

382

### 383 3.3. Seasonal variability and trend of the environmental variables

384 Over the six environmental variables considered, only the depth of the isotherm 20°C ( $D_{\text{iso}20}$ ) was  
 385 found to increase significantly over the two last decades (Fig. 4; Table S2,  $p < 0.05$ ). The five other  
 386 variables (SST, Chl-a, MLD, NPP and  $D_{\text{iso}12}$ ) remained stable between 2001 and 2018 (all  $p > 0.05$ ).  
 387 All physical variables showed strong seasonality over our study period.



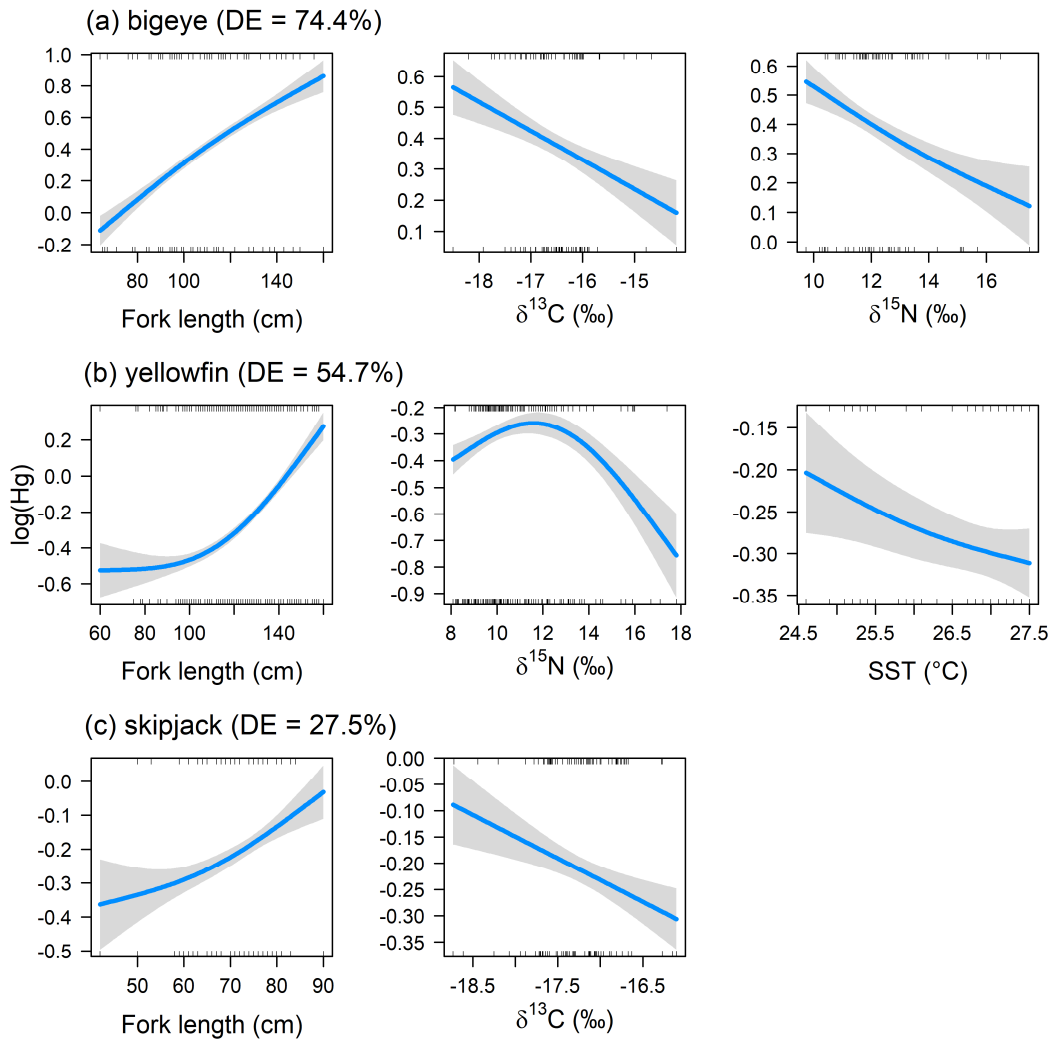
390 Figure 4: Seasonal variations of 6-month averages of oceanic variables in the New Caledonia-Fiji region. a) sea surface  
 391 temperature ( $^{\circ}\text{C}$ ), b) mixed layer depth (m), c) Chl-a ( $\text{mg}\cdot\text{m}^{-3}$ ), d) net primary production ( $\text{mg C}\cdot\text{m}^{-2}\cdot\text{day}^{-1}$ ), e) depth of  
 392 isotherm  $20^{\circ}\text{C}$  (m), f) depth of isotherm  $12^{\circ}\text{C}$  (m), and g) oceanic Niño index (in red: El Niño event; in blue: La Niña

393 event). The red dots represent the months with tuna samples. The grey lines and shadows give respectively the monthly  
394 means and standard deviations over the 6-month period. The black regression line represents the significant temporal  
395 trend.

396

### 397 **3.4. Drivers of the inter-annual variability of tuna Hg content**

398 For bigeye, yellowfin and skipjack respectively, the best models explained 74.4, 49.4 and 27.5 % of  
399 deviance for log(Hg) and 29.3, 16 and 14.2% of deviance for standardized Hg concentrations (Table  
400 2; Figs. 5 & S2). For the three species, fish length appeared as the best stand-alone predictor of  
401 log(Hg), explaining 61.8, 49.4 and 21.3 % of the deviance for bigeye, yellowfin and skipjack  
402 respectively. Considering standardized Hg concentrations (i.e. residuals from the length-based Hg  
403 models), muscle  $\delta^{15}\text{N}$  values were found to be the best stand-alone predictor of Hg distribution for  
404 bigeye and yellowfin; but they were not selected in skipjack's best model. Generally, Hg  
405 concentrations were found to increase with decreasing  $\delta^{15}\text{N}$  values.  $\delta^{13}\text{C}$  values were significant in  
406 the best models of both bigeye and skipjack with lower Hg concentrations related to decreasing  $\delta^{13}\text{C}$   
407 values. SST was selected in the best model of yellowfin only, with response curve predicting lower  
408 Hg concentrations when SST increased. No other oceanographic variables were selected in the  
409 optimal models.



410

411 Figure 5: Results of the optimal generalized additive models (GAM) predicting log(Hg) in a) bigeye, b) yellowfin, and c)  
 412 skipjack. The blue lines give the expected values while the grey bands show the confidence interval for the expected  
 413 value. The ticks at the top and the bottom are the observed values' position associated respectively to positive and  
 414 negative model residuals. DE: deviance explained.

415

416 Table 2: Results of the optimal generalized additive models (GAM) predicting log(Hg) and standardized Hg  
 417 concentrations in bigeye, yellowfin and skipjack. n: number of tuna individuals; DE: deviance explained; *p*-value:  
 418 statistics of the models

Species	Response variable	Explanatory variables	DE (%)	<i>p</i> -value
bigeye	log(Hg)	Length	61.8	<0.001
		$\delta^{13}\text{C}$	11.4	<0.001
		$\delta^{15}\text{N}$	10.5	<0.001



		Length + $\delta^{13}\text{C}$ + $\delta^{15}\text{N}$	74.4	
	standardized Hg	$\delta^{15}\text{N}$	17	<0.001
		$\delta^{13}\text{C}$	8.6	<0.001
		$\delta^{15}\text{N}$ + $\delta^{13}\text{C}$	29.3	
yellowfin	log(Hg)	Length	49.4	<0.001
		$\delta^{15}\text{N}$	13.2	<0.001
		SST	0.34	0.027
		Length + $\delta^{15}\text{N}$ + SST	54.7	
		$\delta^{15}\text{N}$	12.4	<0.001
	standardized Hg	SST	2.31	<0.001
		$\delta^{15}\text{N}$ + SST	16.0	
skipjack	log(Hg)	Length	21.3	<0.001
		$\delta^{13}\text{C}$	9.3	<0.001
		Length + $\delta^{13}\text{C}$	27.5	
	standardized Hg	$\delta^{13}\text{C}$	14.2	<0.001

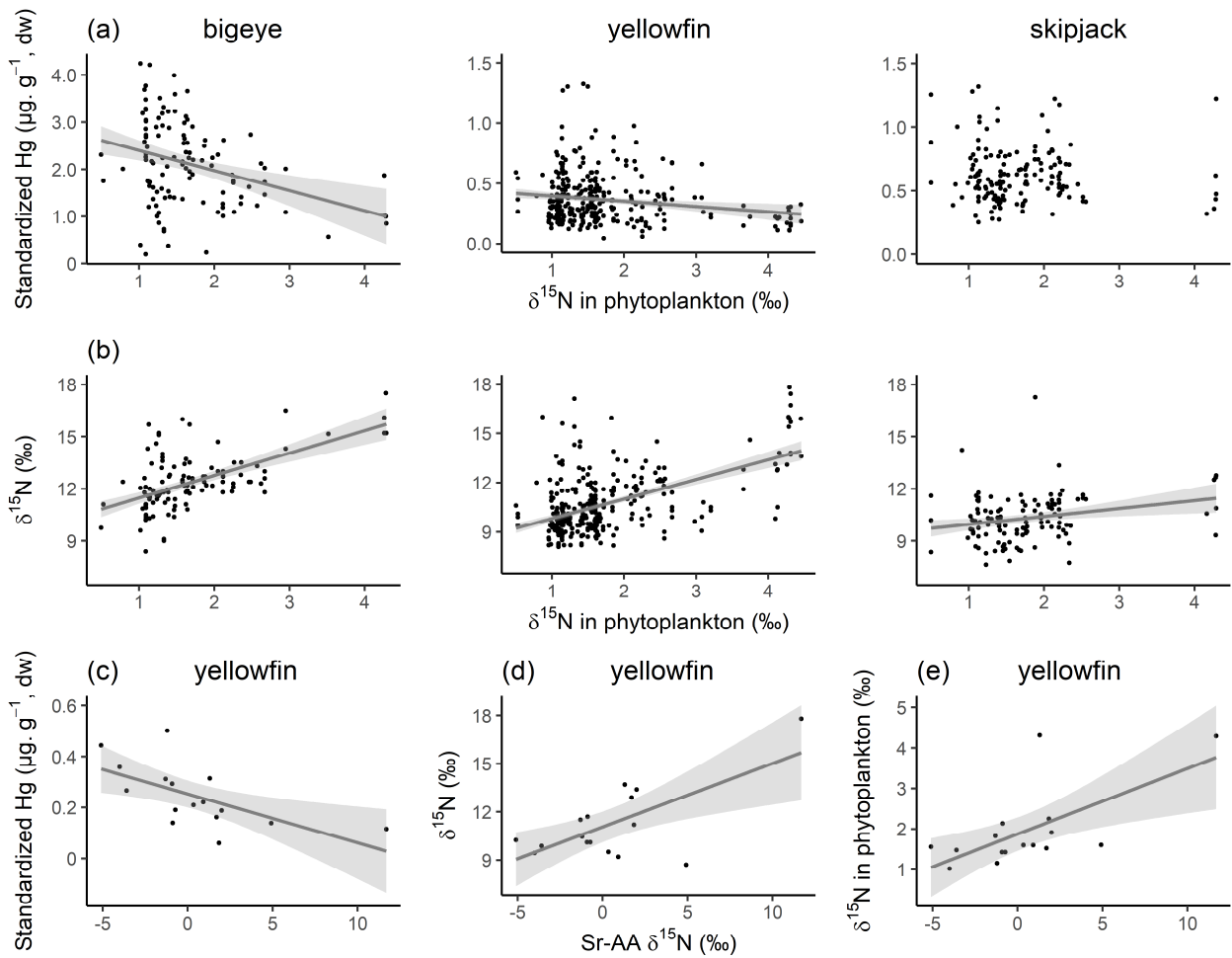
419

420

### 421 3.5. Mercury concentrations and tuna $\delta^{15}\text{N}$ values in relation to baseline processes

422 Standardized Hg concentrations and estimated phytoplankton  $\delta^{15}\text{N}$  values were negatively correlated  
423 for bigeye and yellowfin ( $p < 0.05$ ) while no significant linear relationship was found for skipjack  
424 (Fig. 6a). Conversely, tuna muscle  $\delta^{15}\text{N}$  values were positively correlated to phytoplankton  $\delta^{15}\text{N}$   
425 estimates for the three species (Fig. 6b,  $p < 0.05$ ). For yellowfin samples analysed in AA-CSIA,  
426 standardized Hg was found to be negatively correlated to source amino acids (Sr-AA)  $\delta^{15}\text{N}$  values,  
427 while both tuna muscle and estimated baseline  $\delta^{15}\text{N}$  values were positively correlated to Sr-AA  $\delta^{15}\text{N}$   
428 values (Figs. 6c, 6d & 6e, all  $p < 0.05$ ).

429



430

431 Figure 6: Relationships between a) standardized Hg concentrations ( $\mu\text{g}\cdot\text{g}^{-1}$ , dw) and estimated baseline phytoplankton  
 432  $\delta^{15}\text{N}$  values (‰); and b) tuna muscle  $\delta^{15}\text{N}$  values (‰) and estimated baseline phytoplankton  $\delta^{15}\text{N}$  values (‰). In selected  
 433 yellowfin samples analysed for AA-CSIA, relationships between c) standardized Hg concentrations ( $\mu\text{g}\cdot\text{g}^{-1}$ , dw) and Sr-  
 434 AA  $\delta^{15}\text{N}$  values; d) muscle  $\delta^{15}\text{N}$  values (‰) and Sr-AA  $\delta^{15}\text{N}$  values (‰); and e) estimated baseline phytoplankton  $\delta^{15}\text{N}$   
 435 values (‰) and Sr-AA  $\delta^{15}\text{N}$  (‰). The lines represent the significant linear relationships between the two variables and  
 436 the grey bands show the confidence intervals.

## 437 **4. Discussion**

438 We report the first long-term temporal study of Hg concentrations in tropical commercial tuna from  
439 the south western Pacific Ocean. Contrary to existing temporal studies of tuna Hg content in the  
440 north Pacific Ocean (Hawaii, Drevnick et al., 2015) and in the north western Atlantic Ocean (Lee et  
441 al., 2016), our study revealed the absence of a significant long-term trend of Hg concentrations in  
442 tuna from the New Caledonia-Fiji region during the last two decades. Strong inter-annual variability  
443 of Hg concentrations was however found in all three tuna species and was mainly related to the  
444 variability in tuna sampled lengths among years and to biogeochemical processes occurring at the  
445 primary producer level.

446

### 447 **4.1. Mercury concentrations in tropical tuna**

448 Among the three species, only bigeye tuna exhibited Hg concentrations exceeding the food safety  
449 guideline of  $1 \mu\text{g}\cdot\text{g}^{-1}$  (wet weight) (WHO and UNEP Chemicals, 2008), representing 32% of the  
450 sampled specimens. Overall, most of these individuals (86 %) were bigger than 110 cm. A few  
451 yellowfin were also above the food safety guideline but represented only 0.6% of the dataset. This  
452 illustrates the need to consider both tuna species and fish size when addressing recommendations in  
453 terms of food security regarding Hg content.

454 Relative differences of Hg concentration between the three studied tuna species in the south western  
455 Pacific Ocean were similar to those reported in the north eastern and north western Pacific Ocean  
456 (Blum et al., 2013; Choy et al., 2009; García-Hernández et al., 2007; Yamashita et al., 2005). The  
457 highest Hg content in bigeye compared to the two other species is presumably the result of three  
458 confounding factors: a higher TP for this species, a deeper vertical habitat facilitating its access to  
459 mesopelagic prey with enhanced Hg concentrations, and a longer lifespan (Choy et al., 2009;  
460 Houssard et al., 2019). The significant differences between species muscle  $\delta^{15}\text{N}$  values could indeed

461 suggest a slightly higher TP for bigeye compared to yellowfin and skipjack, as reported in the eastern  
462 Atlantic and the western Indian oceans (Sardenne et al., 2019, 2016). On the other hand, these  
463 differences of  $\delta^{15}\text{N}$  values could also reflect distinct foraging habitat with bigeye occupying deeper  
464 habitats where prey are characterized by higher  $\delta^{15}\text{N}$  values due to baseline effects (Hannides et al.,  
465 2013).

466

#### 467 **4.2. Decadal stability of tuna mercury concentrations**

468 Our study revealed no significant long-term trend of Hg concentrations between 2001 and 2018 in  
469 the New Caledonia-Fiji region despite significant inter-annual variability (discussed separately  
470 below). As samples were collected opportunistically onboard fishing boats, it was not possible to  
471 perform our temporal analysis on the same number of samples for each species. The most complete  
472 and continuous yellowfin dataset illustrates the absence of a time trend over the 18 years, and similar  
473 finding applies to both skipjack and bigeye tuna.

474 Our observed stable long-term trends contrast with the estimated Hg increases of 3.8% annually  
475 reported in yellowfin tuna caught near Hawaii from 1971 to 2008 (Drevnick et al., 2015) and the  
476 mean annual decreasing rate of 19% from the 1990s to the early 2000s found in the Atlantic bluefin  
477 tuna from the north western Atlantic Ocean (Lee et al., 2016). In this later study, the authors  
478 suggested that lower Hg concentrations could be related to the reduction of anthropogenic Hg  
479 emission in North America, implying a direct link between Hg anthropogenic fluctuations and tuna  
480 Hg concentrations.

481 Our findings seem to confirm the fact that Hg bioaccumulation in fish do not necessarily follow the  
482 global suspected increase of anthropogenic Hg emissions to the atmosphere and instead suggest  
483 regional differences in oceanic Hg loads. The lack of long-term trend could be explained in part by  
484 the remoteness of our region, similar to most of the south western Pacific Ocean, which has low

485 anthropogenic emissions and negligible loadings from anthropogenic sources on a total global  
486 emissions scale (Horowitz et al., 2017; UN Environment, 2019). The absence of detectable decadal  
487 trend in tuna Hg concentrations in this region mirrors the absence of consistent temporal trend in  
488 atmospheric Hg concentrations in the southern hemisphere (Slemr et al., 2020).

489 Our contrasted results with the two other temporal studies of Hg concentrations in tuna, located in  
490 the northern hemisphere, could be also related to distinct methodological approaches used to  
491 investigate the temporal trend over time. Drevnick et al. (2015) performed their temporal analysis on  
492 distinct, and highly heterogeneous datasets covering only three years (1971, 1998 and 2008) over a  
493 long study period (37 years), and with a limited number of individuals for the most recent period  
494 (n=14 in 2008). Conversely, we used a quasi-continuous long-term dataset analysed in the same  
495 laboratory from 2001 to 2018 with a larger sample size and more powerful statistical tools. Finally,  
496 unlike our study, no ecological proxies were available in these two other temporal studies of Hg  
497 content; therefore, it was not possible to investigate the potential confounding ecological  
498 contribution to the decreasing or increasing long-term trends of tuna Hg concentrations.

499

### 500 **4.3. Drivers of temporal variability of tuna Hg concentrations**

501 For the three tuna species, body length of sampled fish appeared to be the most important driver of  
502 inter-annual variability of Hg concentrations with highest and lowest Hg concentrations in the time  
503 series related to larger and smaller fish respectively (Figs. 2a, 2b & 2c). This reflects the well-known  
504 bioaccumulative properties of MeHg in organisms (Adams, 2004; Cai et al., 2007; Houssard et al.,  
505 2019) and is mainly related to the variability in tuna sampled sizes among years. Therefore, length-  
506 standardized Hg concentrations are important to use when investigating factors governing Hg  
507 bioaccumulation in tuna. This strong relationship between length and Hg content (61.8%, 49.4% and  
508 21.3 % for bigeye, yellowfin and skipjack respectively) explains the relatively low scores of our

509 modelling approach for length-standardized Hg concentration as most of the variation is already  
510 explained by fish length. Furthermore, for yellowfin and skipjack, where the variance of bulk Hg  
511 concentrations is lower compared to bigeye, by extracting the residuals from the length-based Hg  
512 models to calculate standardized Hg concentrations, we consequently reduced these variances even  
513 more and therefore, the variability to be explained in our GAMs. Considering the ontogenetic dietary  
514 shift in bigeye and yellowfin (Sardenne et al., 2016), fitting GAMs on distinct fish length classes  
515 (e.g. small  $\leq 100$  cm vs large tuna  $> 100$  cm) would have been interesting to investigate differences  
516 among the main processes governing Hg methylation and MeHg bioaccumulation. Unfortunately,  
517 not enough data were available to do so.

518 Considering standardized Hg concentrations (i.e. residuals from length-based Hg models), inter-  
519 annual variability of Hg concentrations in bigeye and yellowfin best correlated with muscle  $\delta^{15}\text{N}$   
520 values, and our predictions showed generally higher Hg concentrations associated with lower muscle  
521  $\delta^{15}\text{N}$  values. In the literature, most studies found that Hg is strongly correlated to organism  $\delta^{15}\text{N}$   
522 values, yet through a positive relationship, reflecting Hg biomagnification along the pelagic food  
523 web (Cai et al., 2007; Teffer et al., 2014). Nevertheless, when exploring muscle  $\delta^{15}\text{N}$  values at the  
524 species level, it is important to keep in mind that they result from trophic dynamics along the food  
525 web, but also from biogeochemical processes at the base of the ecosystem (baseline effects). Here,  
526 muscle  $\delta^{15}\text{N}$  values were positively correlated to estimated baseline phytoplankton  $\delta^{15}\text{N}$  values and  
527 Sr-AA  $\delta^{15}\text{N}$  values, used to track baseline changes in nutrient sources and uptake. Therefore,  
528 temporal variations of muscle  $\delta^{15}\text{N}$  values would predominantly reflect changes at the base of the  
529 food web. This could be related to the high levels of diazotrophy well documented in our study  
530 region (Bonnet et al., 2017; Garcia et al., 2007; Shiozaki et al., 2014) and characterized by low POM  
531  $\delta^{15}\text{N}$  values ( $\sim 1$  ‰). Atmospheric nitrogen deposition from pollution (which typically has  $\delta^{15}\text{N}$   
532 values ranging from -7 to 0 ‰) could also explain the low basal  $\delta^{15}\text{N}$  values; yet as nitrogen  
533 emissions are supposed to be low in our study area compared to the north Pacific Ocean, this

534 phenomenon is likely of less importance compared to diazotrophy (Gobel et al., 2013; Reay et al.,  
535 2008). Thereby, the response curves associated with muscle  $\delta^{15}\text{N}$  values (i.e. higher Hg  
536 concentrations associated with lower  $\delta^{15}\text{N}$  values and thus with higher diazotrophy) in our models  
537 may suggest that the nitrogen cycle and/or diazotrophy (i.e. baseline effects) are likely to drive Hg  
538 concentrations in the upper part of the oceanic pelagic food web, probably triggering Hg net  
539 methylation and bioavailability at the base of the food web during diazotroph blooms and/or their  
540 resulting remineralization. This finding is in line with the recent study by Wu et al. (2019) who  
541 revealed in a meta-analysis that bioconcentration (i.e. MeHg transfer from water into the base of the  
542 food web) was a better descriptor of fish MeHg concentrations than biomagnification in pelagic food  
543 web. The influence of the nitrogen cycle and/or diazotrophy on Hg concentrations seems to be also  
544 confirmed by the negative correlations found in this study between standardized Hg concentrations  
545 and both Sr-AA  $\delta^{15}\text{N}$  values and estimated baseline  $\delta^{15}\text{N}$  values.

546 The community structure and growth of primary producers, as inferred by tuna  $\delta^{13}\text{C}$  values, also  
547 seems to influence the observed inter-annual variability of Hg concentrations, particularly for both  
548 bigeye and skipjack tuna, with Hg concentrations predicted to decrease with increasing  $\delta^{13}\text{C}$  values.  
549 Here,  $\delta^{13}\text{C}$  values were the only ecological parameter showing a significant decline between 2001  
550 and 2018. The same trends were found at the global scale and were attributed to a potential global  
551 shift of the phytoplankton community structure and/or physiology (Lorrain et al., 2020). The lack of  
552 a corresponding long-term Hg trend in our study may suggest that the carbon cycle is likely to  
553 impact Hg fate in oceans and tuna but to a lesser extent than the nitrogen cycle, and in particular  
554 diazotroph blooms.

555 As dinitrogen fixers generally grow well in warm and stratified conditions, those conditions, more  
556 than the nitrogen cycle itself, could be linked to the Hg cycle. Recent model predictions made on  
557 Atlantic bluefin tuna show that increasing tissue Hg concentrations were related to rising seawater  
558 temperatures (Schartup et al., 2019). Furthermore, experimental studies on estuarine and freshwater

559 fish have found that warmer temperatures enhanced MeHg bioaccumulation (Dijkstra et al., 2013;  
560 Maulvault et al., 2016). However, here we find SST was selected in the optimal model for yellowfin  
561 only, showing an opposing response curve to the above hypothesis, i.e. lower Hg concentrations in  
562 tuna related to higher SST. Our response curve for yellowfin is in agreement with Houssard et al.  
563 (2019) where Hg content was found to decrease spatially with increasing SST. Considering the  
564 potential impact of stratification, the depth of isotherm 12°C ( $D_{iso12}$ , which can be used as a proxy of  
565 the thermocline and thus stratification) was not selected in any of our optimal models. In Houssard et  
566 al. (2019),  $D_{iso12}$  was found as the second main driver (after fish length) of the spatial variability of  
567 tuna Hg content with Hg concentrations enhanced in regions characterised by deeper thermoclines.  
568 These distinct results may suggest that the stand-alone temporal variability of the thermocline depth  
569 is not responsible for the inter-annual variations of Hg concentrations in tuna muscle. While  
570 discussing the impact of these environmental variables, it is worthwhile mentioning that we worked  
571 at a sub-regional scale where temporal variability is low compared to large spatial variability  
572 investigated in these other studies. For example, the ranges of our physical variables, especially for  
573  $D_{iso12}$  (428 – 451 m), were reduced compared to the ones investigated in Houssard et al. (2019)  
574 ( $D_{iso12}$ : 200 – 460 m), which can explain their limited impact in our models.

575 Including complementary Hg data from the environment (i.e. Hg concentrations in seawater and total  
576 gaseous Hg) in our modelling approach could have improved our model scores and help  
577 characterizing in what extent Hg levels in tuna reflect Hg levels in water or in the atmosphere.  
578 Unfortunately, such data are scarce and no time series over the two last decades were available in our  
579 study area.

580

#### 581 **4.4. Implications for global-scale monitoring of mercury in tuna**



582 With the adoption of the Minamata Convention in 2013, governments are asked to control and  
583 reduce anthropogenic Hg emissions, requiring diverse ecosystem and organism studies, as well as  
584 monitoring tools and strategies to track the efficiency of political decisions. Considering that tuna  
585 fisheries are among the world's most important fisheries, with a commercial value estimated at 41  
586 billion \$US/y in 2012 (Macfadyen, 2016), quantifying nutritional risks along with benefits are  
587 becoming important when addressing food and nutritional security. Furthermore, knowing that  
588 oceans have undergone large physical and biogeochemical modifications in the last decades (e.g.  
589 surface warming, acidification, deoxygenation or changes in primary productivity) (Bopp et al.,  
590 2013; Kwiatkowski et al., 2017), it has become necessary to assess the predictive capacity of  
591 environmental variables on Hg methylation and bioaccumulation to understand potential climate-  
592 driven ecological changes.

593 With a continuous and long-term dataset, we revealed for the first time the absence of significant  
594 decadal trend of Hg concentrations in tropical tuna from the New Caledonia-Fiji region. This  
595 contrasts with the two other available temporal studies from distinct areas (Drevnick et al., 2015; Lee  
596 et al., 2016) and thus illustrates the complexity of the Hg cycle and the fact that Hg in tuna does not  
597 necessarily follow the suspected recent increasing Hg trends in global oceans, especially in the  
598 southern hemisphere. Our results however seem consistent with the remoteness of our study region  
599 in terms of low anthropogenic emissions, and thus seem to confirm the hypothesis of distinct  
600 hemispherical ocean patterns of Hg anthropogenic deposition.

601 Strong inter-annual variability was found in the three tuna species and was mainly due to fish length  
602 variability among samples. This suggests the importance of using a fish length-based approach to  
603 address the question of Hg spatial and temporal trends in tuna even at small (e.g. sub-regional)  
604 scales. This includes accounting for different types of fishing gear (e.g. purse seine and longline)  
605 employed since fishing gears are known to preferentially select certain tuna fish sizes.

606 Finally, our novel complementary investigation of baseline and trophic ecological tracers highlights  
607 i) no significant decadal change in tuna TP during this 18 year period, and ii) the influence of  
608 baseline processes for bigeye and yellowfin, related to the nitrogen cycle and/or diazotrophy,  
609 possibly enhancing Hg methylation and/or MeHg bioavailability at the base of the food web.  
610 Knowing that the largest bioaccumulation step of MeHg is likely to occur between the water  
611 compartment and unicellular planktonic organisms - relative to trophic amplification processes -  
612 more attention needs to be paid to ecological and physiological processes occurring at the base of  
613 marine ecosystems to better capture Hg spatio-temporal trends at the top of the ocean food webs.

614 Lastly, our study emphasizes the need for more systematic collection of Hg and stable isotope data in  
615 different marine reservoirs, including iHg in the water column, from both hemispheres to compare  
616 MeHg production, degradation and bioaccumulation in oceans at a global scale.

617 **Declaration of competing interests**

618 The authors declare that they have no known competing financial interests or personal relationships  
619 that could have appeared to influence the work reported in this paper.

620

621

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# Graphical abstract

