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1 **Distribution of mercury species in different tissues and trophic levels**
2 **of commonly consumed fish species from the south Bay of Biscay**
3 **(France)**
4

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20 **Abstract**

21 Mercury (Hg) is a contaminant of global concern in marine ecosystems, notably due to its ability to
22 accumulate and concentrate in food webs. Concentrations of total mercury (THg), methylmercury
23 (MeHg) and inorganic mercury (IHg) were assessed and compared in different tissues (liver, muscle,
24 and gonads) of three common fish species (hake *Merluccius merluccius*, red mullet *Mullus surmuletus*,
25 and sole *Solea solea*) from the continental shelf from the southern part of the Bay of Biscay. Several
26 studies investigated Hg concentration in fish muscle, but few assessed concentrations in other organs,
27 despite the importance of such data to understand contaminant organotropism and metabolization.
28 Results showed that trophic position and feeding habitat are required to understand the variability of
29 Hg concentration in muscle between fish species. In addition, high MeHg/THg ratio in muscle could
30 be explained by the predatory behavior of the studied fish species and the biomagnification of this Hg
31 species within the food web, MeHg. Despite differences between species, Hg concentration was
32 always higher in muscle (from 118 ± 64 to 338 ± 101 ng g⁻¹ w.w.) and liver (from 122 ± 108 to $271 \pm$
33 95 ng g⁻¹ w.w.). These results can be related to physiological processes especially the MeHg
34 detoxification strategies.

35

36 **Keywords:** Methylmercury, Contamination; Organotropism; Bioaccumulation; Teleostei

37

38 **Introduction**

39 Contaminants in marine environments are of high concern for both ecological and human health
40 reasons. Within the Marine Strategy Framework Directive (MSFD), the European Union fosters the
41 achievement of good environmental status of marine waters by 2020. Two MSFD descriptors deal
42 with contaminants: descriptor 8 aims at limiting contaminant levels below adverse effect for
43 ecosystem functioning while descriptor 9 aims at limiting contaminant levels below sanitary threshold,
44 i.e. below adverse levels for human consumers (EC, 2008). Among the contaminants of interest,
45 mercury (Hg) is a non-essential heavy metal, recognized as hazardous for living organisms. Hg is
46 emitted in the environment from natural (e.g. volcanic eruptions) and anthropogenic sources (e.g.
47 burning of fossil fuels or industry) in a gaseous form. It became widespread in the aquatic ecosystems
48 mainly through atmosphere due to its high volatility. After deposition as inorganic species (IHg), it
49 can be converted into its organic methylated form - namely methylmercury (MeHg)- by bacterial
50 activities (Villar et al., 2020). All Hg species (organic and inorganic) are bioaccumulated in organisms
51 but only MeHg is well-known for its biomagnification property, namely the trend for increasing
52 concentrations along food webs (Baeyens et al., 2003; Bryan et al., 1979). Because feeding is the main
53 pathway responsible for Hg transfer in marine organisms (Cresson et al., 2014; Hall et al., 1997), high
54 Hg concentrations are generally observed in high trophic level species such as piscivorous fish species
55 (Cossa et al., 2012), mammals (Brown et al., 2018; Pinzone et al., 2019) or sharks (Endo et al., 2008;
56 McKinney et al., 2016). In addition, MeHg dominates in fish muscle, which may imply a high
57 toxicological hazard for humans, considering that most wild fish species consumed are at high trophic
58 level (Lavoie et al., 2018; Pauly et al., 1998) and that muscle is the main edible fish tissue. Therefore,
59 due to Hg potent high neurotoxic effects for human, Hg concentrations were largely monitored (1) in
60 fish muscle, to document potential health hazard, and (2) in aquatic biota, as to reconstruct Hg
61 pathway along food chains (Lavoie et al., 2013). However, research on other tissues is important to
62 understand Hg major contamination targets. Moreover, Hg species distribution allows to elucidate
63 tissue-specific bioaccumulation pathways and to investigate different behavior for organic and
64 inorganic Hg species, with implications for organisms toxicokinetics (Kasper et al., 2009). In addition

65 to the organism metabolic activity, fish bioaccumulation pattern is a complex process, where Hg
66 concentrations result from combined effect of both abiotic (e.g. habitats of species (Chouvelon et al.,
67 2012; Cresson et al., 2015a)) and biotic factors (e.g. fish growth, trophic functioning of ecosystems
68 (Chouvelon et al., 2018; Cossa et al., 2012; Cresson et al., 2014)). In this context, the aim of this study
69 was to investigate Hg species (MeHg and IHg) distribution among three organs (muscle, liver, and
70 gonad) in three commercial fish species, considering that differences in diet and feeding habitat can be
71 potential drivers of differences.

72 **Material and methods**

73 *Sampling area*

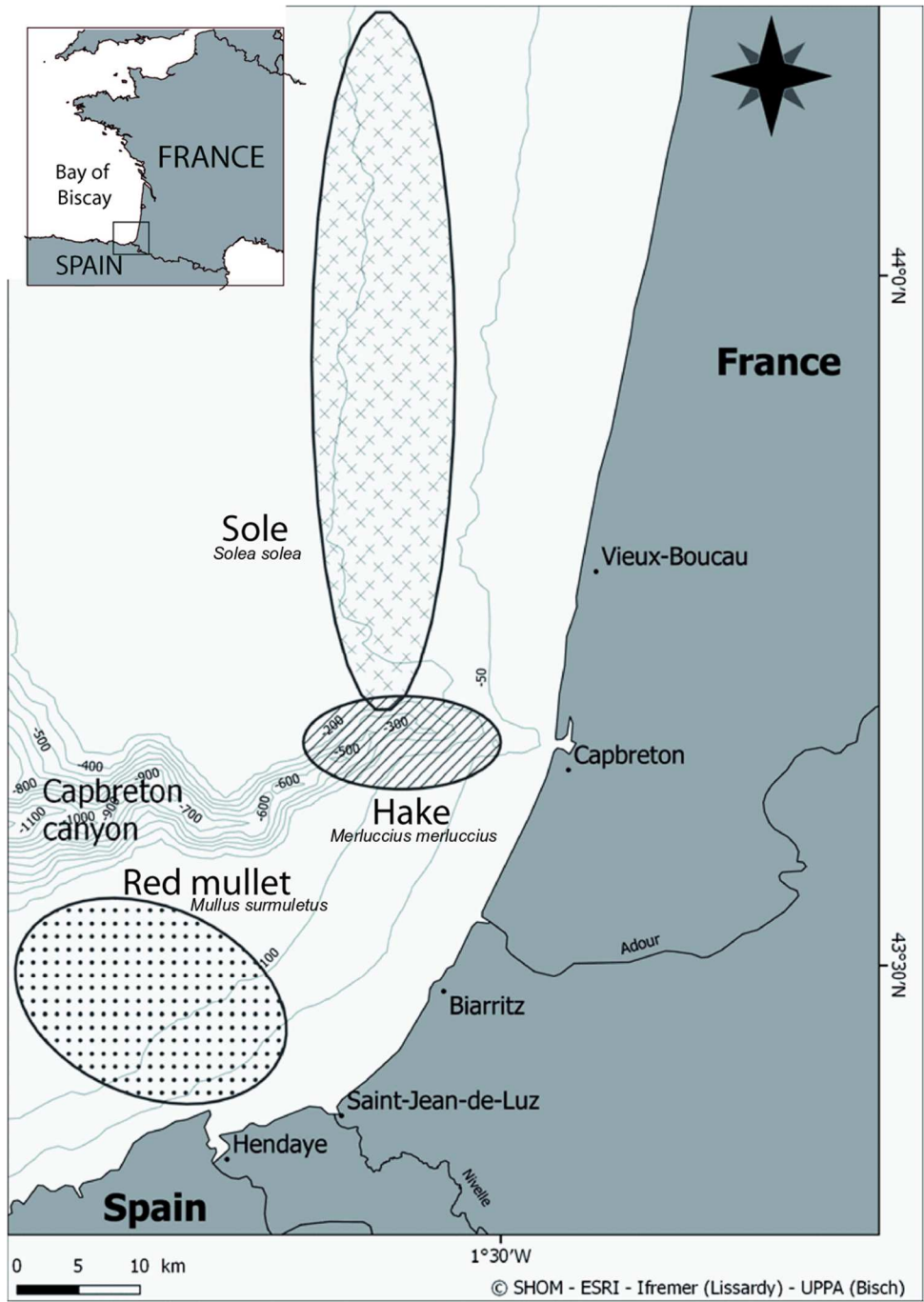
74 Fish were sampled in the southern part of the Bay of Biscay, in the Eastern Atlantic Ocean (Figure 1).
75 This area is characterized by a rocky zone in the south, a canyon in the center and a sandy zone in the
76 north. The Adour, the small Pyrenean and northern Spanish coastal rivers from the Cantabrian chain
77 contribute to the volumes of suspended matter exported to the ocean; the Adour contributes $0.25 \cdot 10^6$
78 tons per year and the Pyrenean coastal rivers $1.9 \cdot 10^6$ tons per year (Jouanneau et al., 1999; Maneux et
79 al., 1999). The hydrodynamics of the Basque plateau is dominated by the processes associated with
80 swells and storms (Uriarte et al., 2004). The southeastern part of the Bay of Biscay is characterized by
81 the presence of a contour current that runs along the edge of the Basque-Cantabrian platform (from
82 west to east), then the edge of the Aquitaine platform (from south to the North). This current results
83 from a contrast of density between the waters of the platforms and the deep waters.

84

85 *Species selection and sampling collection*

86 Three fish species were selected for their commercial interest in the Bay of Biscay, their availability
87 regarding sampling on various habitats characteristics and different feeding habitats. Hake (*Merluccius*
88 *merluccius*) is a demersal species with high commercial interest (19% of the landing value in 2017)
89 within the Bay of Biscay (Ifremer Fisheries Information system, 2018). Canyons areas, especially the
90 continental shelf accretion, promote concentrations of adults in the south Bay of Biscay (Alvarez et al.,
91 2001; Sánchez and Gil, 2000; Woillez et al., 2007). Two common benthic fishes, common sole (*Solea*

92 *solea*) and striped red mullet (*Mullus surmuletus*) representing 6% and 1% of the landing value
93 respectively (Ifremer Fisheries Information system, 2018) were also selected. Sole habitat is fine and
94 muddy sands or vases (Koutsikopoulos and Lacroix, 1992) whereas red mullet lives on sandy,
95 gravelly, rocky, shellfish bottoms (Quéro and Vayne, 1986). A total of 90 individuals of the three
96 species were caught by fishermen in 2017 and 2018: 32 hakes from the continental shelf of the
97 Capbreton canyon, 17 red mullets from the rocky bottom of French Basque coast, and 41 soles caught
98 on the sandy Landes coast (Table 1 & Figure 1). In order to investigate sources of organic matter, the
99 sampling also included zooplankton samples collected with a vertical plankton net (200 µm mesh size)
100 at two stations in the Adour plume (Zooplankton P, 20m depth) and in the head of the Capbreton
101 canyon (Zooplankton C, 120m depth). Surface water samples (-1 m) were also collected at these two
102 points and in the Adour estuary with a 20 L Niskin bottle, and directly filtered through pre-combusted
103 (4 h–450 °C) GF/F filters (47 mmØ) for C and N isotopic analyses of suspended particulate organic
104 matter (POM). Prior to analyses, filters were dried overnight at 50 °C and were decarbonated by
105 contact with HCl vapor (8 h), following a classical protocol (*e.g.* Dubois et al., 2012; Liénart et al.,
106 2016; Savoye et al., 2012). In order to limit the effect of several biotic factors such as fish age and/or
107 length (Cresson et al., 2015a), individuals were caught in a limited range of fish size, corresponding to
108 the age of the first reproduction. Hake breeds around 40 cm for males (2 years old) and around 50-60
109 cm (3-4 years old) for females. For sole, the first sexual maturity usually occurs at the age of 2 or 3
110 years and around 25 to 30 cm long (Koutsikopoulos and Lacroix, 1992). In the Bay of Biscay, sizes of
111 first sexual maturity for red mullet were around 16 cm and 18 cm for males and females, respectively
112 (Dorel, 1986). The size and sex effects on Hg burden have been tested.



113
 114 Figure 1: Map of sampling areas for the three fish species studied.

115 *Mercury species analysis*

116 For all individuals, after being weighted and measured, white muscle (without skin), liver and gonad
 117 were dissected out. Hg was measured in all tissues, while isotopic analyses were performed in muscle
 118 only, as this tissue is the most classical and the most suited to estimate fish average trophic position.
 119 All samples were then freeze-dried, homogenized and grinded.

120 Hg species were measured by capillary gas chromatography (Focus GC, Thermo Electron) connected
121 to an inductively coupled plasma mass spectrometer (ICPMS X2 series, Thermo Electron).
122 Methodology and analytical set-up for the GC-ICP-MS for Hg speciation analysis are detailed in
123 Monperrus *et al.* (2005). Briefly, about 100 mg of homogenized dry tissue were digested with 5 ml
124 TMAH (Tetramethylammonium hydroxide) under a microwave field and then centrifuged to remove
125 solid particles. The supernatant, stored at 4°C, was then submitted to derivatization using sodium
126 tetraethylborate (3%). Quantification of Hg species was performed by species specific isotope dilution,
127 by adding the appropriate amount of isotopically enriched Hg standards (¹⁹⁹IHg and ²⁰¹MeHg), and by
128 applying isotope pattern deconvolution for data processing (Rodríguez Martín-Doimeadios *et al.*,
129 2004). THg concentrations are equal to the sum of IHg and MeHg concentrations. %MeHg was
130 calculated as the ratio between MeHg and THg. Analytical performances were checked by blanks and
131 by Certified reference materials (CRMs) DOLT-4 (Dogfish liver) and TORT-2 (Lobster
132 hepatopancreas) from the National Research Council of Canada (CNRC) and BCR464 (Tuna fish
133 muscle) from the Institute for Reference Materials and Measurements (IRMM). Limits of
134 quantification were 1.2 ng g⁻¹ for Hg species. Hg species concentrations were expressed relatively to
135 dry weight. The conversion of Hg concentrations in wet weight (w.w.) was achieved with a conversion
136 factor obtained by weighing the mass difference before and after freeze-drying for all tissues.

137 *Carbon and nitrogen stable isotope analysis*

138 Muscle stable isotopic ratios were measured with a Thermo Scientific® Delta V Advantage mass
139 spectrometer coupled to a Thermo Scientific® Flash EA1112 elemental analyzer following routine
140 protocol of the Plateforme Spectrometrie Ocean ([https://www-ium.univ-brest.fr/delta-v-plus-ea-](https://www-ium.univ-brest.fr/delta-v-plus-ea-gb/?lang=en)
141 [gb/?lang=en](https://www-ium.univ-brest.fr/delta-v-plus-ea-gb/?lang=en)) and used in previously published paper (*e.g.* Mille *et al.*, 2020; Timmerman *et al.*, 2020).
142 For suspended POM samples, carbon and nitrogen isotopic compositions were analyzed using an
143 isotope ratio mass spectrometer (IRMS; Isoprime, GV Instruments®) coupled with an elemental
144 analyzer (ThermoFisher® Scientific Flash 2000). All isotopic results are presented in the classical δ
145 notation, $\delta X = \left(\frac{R_{sample}}{R_{standard}} - 1 \right) \times 10^3$ where X is ¹³C or ¹⁵N and R the ratio between heavy and light
146 isotopes. Standard is Vienna Pee Dee Belemnite for $\delta^{13}\text{C}$ and atmospheric nitrogen for $\delta^{15}\text{N}$. Carbon

147 and nitrogen concentration were measured with the elemental analyzer and used to calculate molar
148 C/N ratios, classical proxy of lipid content in fish tissue for isotopic analyses. High C/N ratios are
149 usually interpreted as high lipid content that bias isotopic measurement. All C/N ratios of fish samples
150 were lower than 3.5 classically considered as the threshold values requiring lipid correction (Sweeting
151 et al., 2006). Based on replicate measurements of internal laboratory standards, the analytical
152 associated uncertainties were 0.13‰ and 0.12‰ for $\delta^{13}\text{C}$ and $\delta^{15}\text{N}$ respectively, and ≤ 0.05 for C/N
153 ratios.

154 *Statistical analysis*

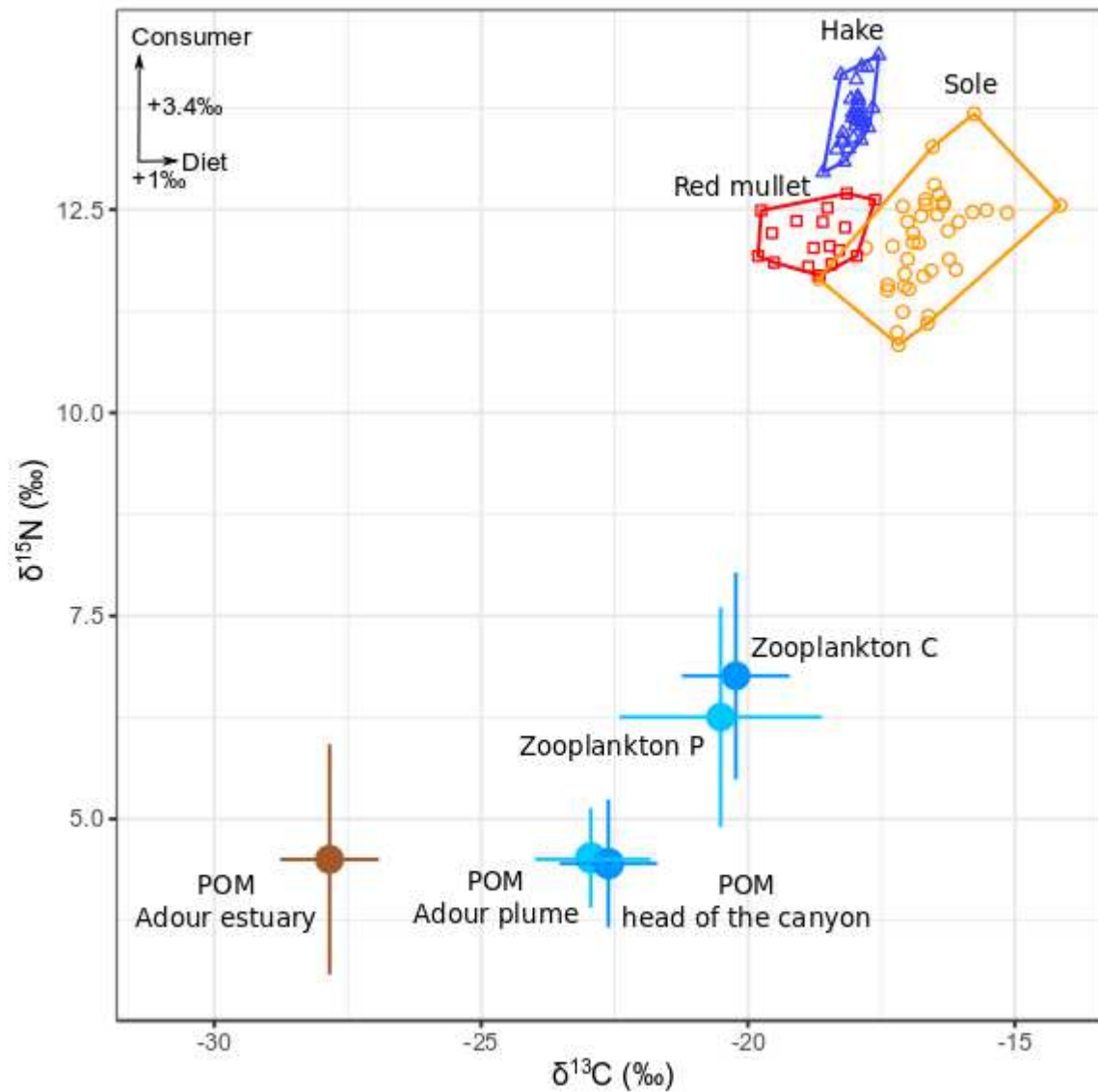
155 For the three fish species, differences in THg concentration and proportion of MeHg (%MeHg) among
156 species were detected with Permutational multivariate analysis of variance (PERMANOVA). At the
157 intra-specific level, the Hg species distribution between organs was investigated with PERMANOVA
158 realized on Hg species concentrations. All statistical analyses were performed to the error threshold of
159 5%, using the “vegan” (Oksanen et al., 2013) package in the statistical environment R Core Team
160 (2018).

161

162 **Results**

163 *Feeding ecology*

164 Sole had the highest $\delta^{13}\text{C}$ values ($-16.80 \pm 1.23\text{‰}$), red mullet the lowest ($-18.72 \pm 0.64\text{‰}$) and hake
165 presented intermediate values ($-17.99 \pm 0.21\text{‰}$) (Figure 2 & Table 1). Values for zooplankton were
166 lower (Zooplankton G: $-20.56 \pm 0.90\text{‰}$; Zooplankton P: $-20.28 \pm 1.18\text{‰}$) followed by the POM
167 values measured in surface waters (head of the canyon: $-22.59 \pm 0.78\text{‰}$; Adour plume: $-22.87 \pm$
168 0.92‰ and Adour estuary: $-27.84 \pm 0.93\text{‰}$). Regarding $\delta^{15}\text{N}$, ratios were higher for hake ($13.64 \pm$
169 0.34‰) than for the two other species ($12.50 \pm 0.31\text{‰}$ and $12.10 \pm 0.58\text{‰}$ for red mullet and sole
170 respectively) and for zooplankton (Zooplankton C: $6.68 \pm 1.29\text{‰}$; Zooplankton P: $6.44 \pm 1.23\text{‰}$) and
171 POM values (head of the canyon: $4.47 \pm 1.00\text{‰}$; Adour plume: $4.56 \pm 0.78\text{‰}$ and Adour estuary: 4.50
172 $\pm 1.42\text{‰}$).



173
 174 Figure 2: Relationships between carbon and nitrogen stable isotopes ratio measured in the three fish
 175 species (hake *Merluccius merluccius*, red mullet *Mullus surmuletus* and sole *Solea solea*). Signatures
 176 of particle organic matter of the Adour estuary, Adour plume, the head of the Capbreton canyon and
 177 signatures of zooplankton from Adour plume and the head of the Capbreton canyon are added (dot:
 178 mean, error bar: standard deviation). The theoretical isotopic change between a consumer and its diet
 179 ($+1\text{‰}$ for $\delta^{13}\text{C}$, $+3.4$ for $\delta^{15}\text{N}$) is represented in the upper left corner of the plot.
 180
 181

182 Table 1: Biometric fish data: number of samples N, fish total length and isotopic signatures (mean \pm standard deviation and range), and concentrations of total
 183 mercury (THg) and methylmercury (MeHg) according to organs/tissues (mean, standard deviation and range).

Species Diet and habitat	N	Total length (mm)	$\delta^{13}\text{C}$ (‰)	$\delta^{15}\text{N}$ (‰)	C/N	THg (ng g ⁻¹ w.w.)			MeHg (ng g ⁻¹ w.w.)		
						Muscle	Liver	Gonads	Muscle	Liver	Gon
Hake <i>Merluccius merluccius</i> demersal piscivore fish	32	550 \pm 60 412 - 665	-17.99 \pm 0.21 -18.59 - -17.55	13.64 \pm 0.34 12.96 - 14.40	3.15 \pm 0.03 3.08 - 3.24	268 \pm 155 78 - 659	123 \pm 43 50 - 228	57 \pm 27 14 - 133	177 \pm 89 56 - 466	73 \pm 37 29 - 199	41 \pm 11
Red mullet <i>Mullus surmuletus</i> benthic invertivore rocky shore	17	241 \pm 14 225 - 287	-18.72 \pm 0.64 -19.81 - -17.62	12.15 \pm 0.31 11.69 - 12.70	3.19 \pm 0.02 3.15 - 3.22	338 \pm 101 214 - 583	271 \pm 95 178 - 521	87 \pm 23 52 - 134	235 \pm 57 161 - 364	225 \pm 84 152 - 451	67 \pm 44
Common sole <i>Solea solea</i> benthic invertivore sandy-muddy shore	41	307 \pm 26 254 - 360	-16.80 \pm 1.23 -22.99 - -14.15	12.10 \pm 0.58 10.84 - 13.27	3.17 \pm 0.04 3.11 - 3.32	118 \pm 64 38 - 340	122 \pm 108 44 - 543	38 \pm 25 10 - 141	87 \pm 50 27 - 260	66 \pm 48 21 - 244	20 \pm 5

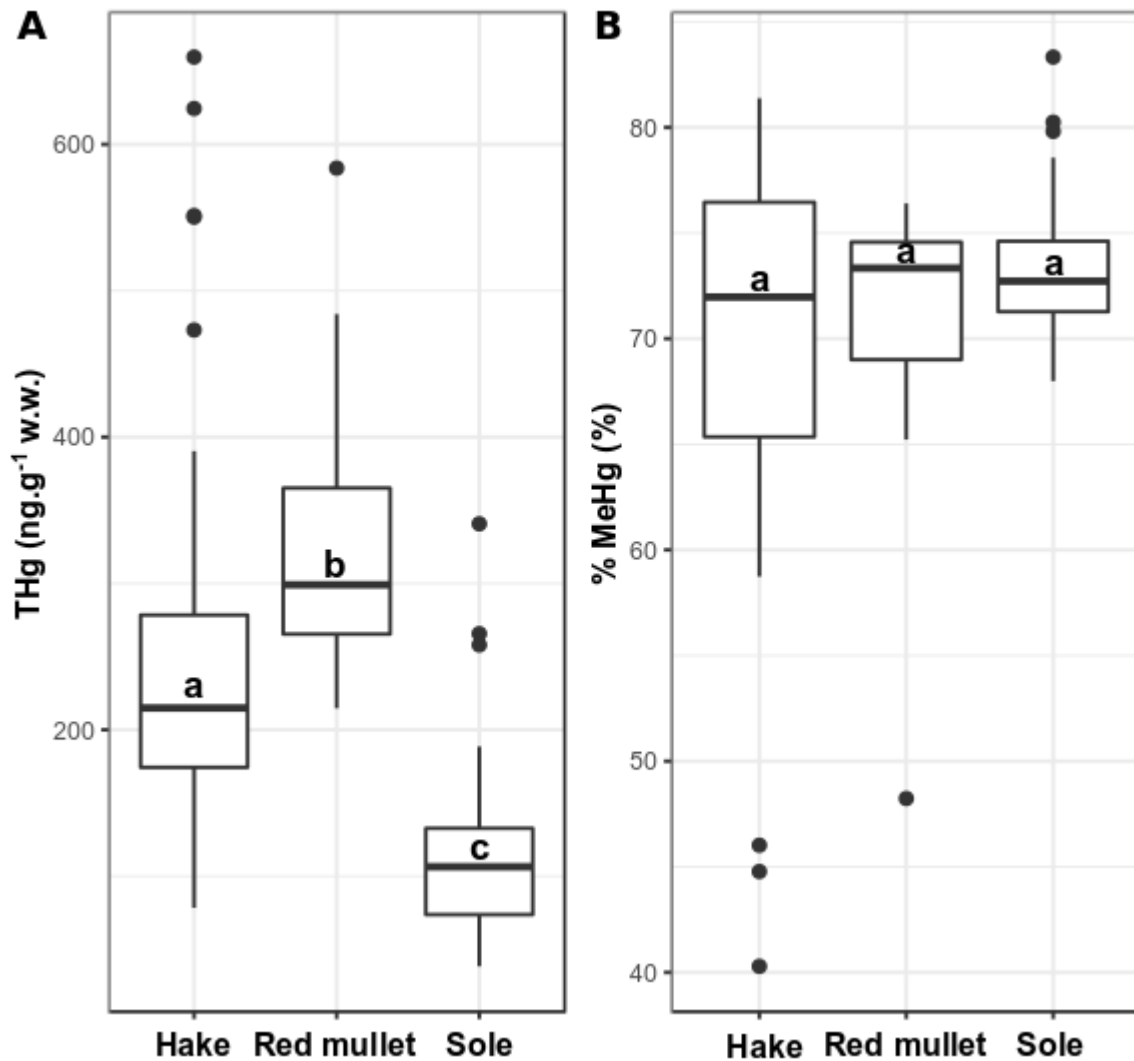
184

185 *Interspecies difference in mercury species concentrations in muscle*

186 In the present study, all individuals, except four hakes, had THg concentrations below the food safety
187 thresholds determined by the European commission regulation [*i.e.* 0.5 $\mu\text{g g}^{-1}$ w.w. for hake and sole,
188 1 $\mu\text{g.g}^{-1}$ w.w. for red mullet (EC, 2006)]. Values above the threshold measured for hakes ranged
189 between 0.55 and 0.66 $\mu\text{g g}^{-1}$ w.w.

190 Hg concentrations in muscle were significantly lower ($F= 30.04$, $p.\text{value} < 0.001$) in sole (118 ± 64 ng
191 g^{-1}) than in hake (268 ± 155 ng g^{-1}) and red mullet (338 ± 101 ng g^{-1}) (Table 1), but %MeHg was
192 comparable between these three species ($\sim 70\%$ on average; $F= 2.79$, $p.\text{value} > 0.05$; Figure 3).

193 Despite the sampling strategy to limit effect of biotic factors, a significant positive linear relationship
194 between fish total length and MeHg in muscle was detected for hake only and a sex effect on Hg
195 concentration was detected for sole only. Detailed results are presented in Table S1 and Figures S1
196 and S2. .



197
 198 Figure 3: Total mercury concentration (A) and proportion of methylmercury (B) measured in muscle
 199 for the three species (hake *Merluccius merluccius*, red mullet *Mullus surmuletus* and sole *Solea solea*).
 200 The bottom and top of the box are the first and the third quartiles of the data distribution, the
 201 horizontal segment is the median, the whiskers represent the most extreme data point within 1.5
 202 interquartile range and black points are data points out of this range. Letters highlight significant
 203 differences between species based on PERMANOVA.

204 *Distribution of mercury species between organs*

205 Among the three organs considered, muscle exhibited higher THg concentration than liver and gonads
 206 for hake and red mullet (Figure 4 & Table 2). Concentration was not different between muscle and
 207 liver for sole, but higher than in the gonads (Figure 4). MeHg concentrations were higher in the
 208 muscle than in the liver and the gonads for the three species. Concentrations were always significantly
 209 lower in the gonads for the three species and higher in the muscle than in the liver, but this difference
 210 is significant for hake only. The pattern was somehow similar for IHg.

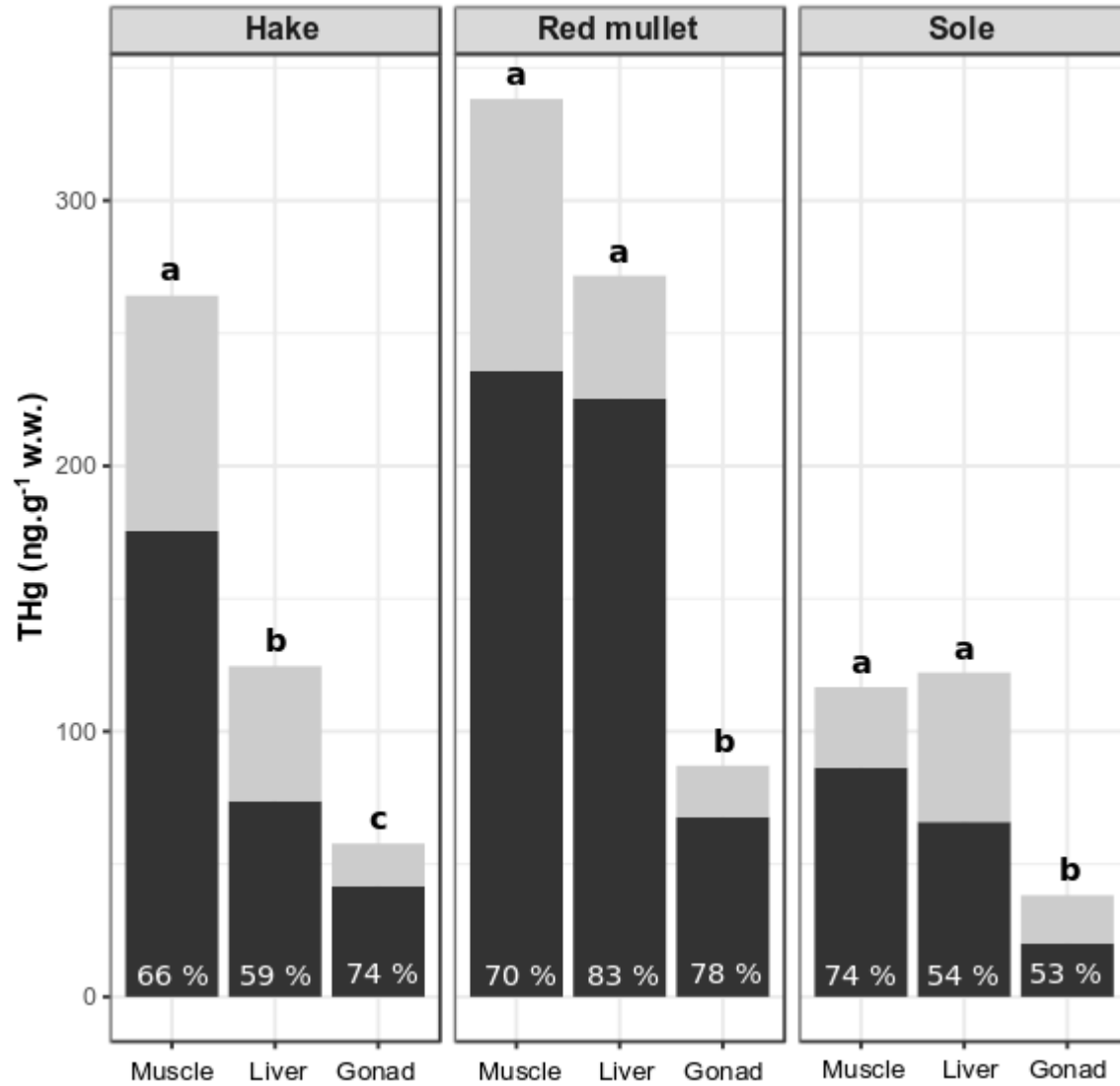
211 %MeHg ratio was organ- and species-specific (Figure 4). In hake, ratios were significantly higher in
 212 gonad (average 74%) than in muscle (average 66%) than in liver (average 59%). For red mullet, the

213 highest %MeHg was measured in liver (average 83%) and in gonad (average 78%), while sole had
214 highest %MeHg in muscle (average 74%).

215 Table 2: Differences in Hg species concentrations according to organs/tissues for the three fish species. Results were obtained with PERMANOVA illustrated
 216 by the F statistic together with the corresponding p.value.

Species	THg	MHg	IHg
Hake <i>Merluccius merluccius</i>	F = 40.15, p.value < 0.001 Muscle > Liver > Gonad	F = 48.38, p.value < 0.001 Muscle > Liver > Gonad	F = 16.13, p.value < 0.001 Muscle > Liver > Gonad
Red mullet <i>Mullus surmuletus</i>	F = 43.48, p.value < 0.001 Muscle > Liver > Gonad	F = 42.28, p.value < 0.001 Muscle = Liver > Gonad	F = 25.17, p.value < 0.001 Muscle > Liver > Gonad
Common sole <i>Solea solea</i>	F = 16.08, p.value < 0.001 Muscle = Liver > Gonad	F = 27.23, p.value < 0.001 Muscle = Liver > Gonad	F = 10.77, p.value < 0.001 Muscle = Liver > Gonad

222



223

224

the three fish species (hake *Merluccius merluccius*, red mullet *Mullus surmuletus* and sole *Solea solea*). MeHg is displayed in black together with its percent

Figure 4: Distribution of Hg species according to organs for

225 relative to THg, and IHg is in grey. Standard deviation are not added for graphical purposes. Significant differences between organs are highlighted with
226 different letters.

227

228 **Discussion**

229 *Influence of diet and feeding habitat on Hg species concentrations in muscle*

230 The present study confirmed that MeHg represent the majority of Hg in fish, consistently with most literature records (Chouvelon et al., 2018; Pethybridge et
231 al., 2010; Polak-Juszczak, 2018). These values can be linked with diet and especially with the trophic level of studied species. The three species are demersal
232 predators (hake) or benthic invertivore (red mullet and sole). Hake diet is mostly based on fish like blue whiting, horse mackerel or sardine. Older individuals
233 can exhibit cannibalism and consume young conspecifics (Mahe et al., 2007; Pinnegar, 2014; Velasco and Olaso, 2000). Red mullet and sole are benthic
234 omnivorous fish consuming small crustaceans, worms and mollusks including small bivalves (Caill-Milly et al., 2017; Le Loc'h et al., 2008). The pattern
235 observed for nitrogen isotopic ratio, with higher values for hake, is thus consistent the expected relative trophic position of the three fish species. Therefore,
236 the species-specific %MeHg observed in this study can be explained by the species diet which, in thus, involves the species specific accumulation of MeHg in
237 muscle. This study highlighted also the biomagnification of MeHg and thus the transfer of this Hg species in the trophic network, resulting in high MeHg
238 concentration in a piscivorous fish species (Cossa et al., 2012). However, the trophic position (estimated by $\delta^{15}\text{N}$ values) is not sufficient to explain the high
239 variability of Hg concentration. Fish position in the water column was also an important factor, since pelagic species presented significantly lower THg
240 concentration in muscle than the benthic species (Chouvelon et al., 2012). This pattern is exemplified here, as red mullet had highest THg concentrations than
241 hake, despite having lower $\delta^{15}\text{N}$ values, consistently with the expected diet of the species. This pattern is even more surprising as hake was caught near the
242 Capbreton canyon, a presumed sink for organic matter, sediment and contaminants especially Hg (Azaroff et al., 2019; Gaudin et al., 2006). This pattern was
243 also observed in the Mediterranean, where fish living in canyons were more contaminated than counterparts living away (Cresson et al., 2016; Koenig et al.,

244 2013). Hake contamination burden can be explained by the fact that all individuals in this study came from the continental shore at the head of the Capbreton
245 canyon that was less contaminated in Hg than offshore (Azaroff et al., 2019). Isotopic values measured for hake are consistent with values previously
246 measured for this species in the Bay of Biscay (Chouvelon et al., 2012; Cossa et al., 2012; Le Loc'h et al., 2008) and with the expected pelagic behavior of
247 this species. It may thus be assumed that they poorly use benthic organic matter from the canyons, but fed above on the pelagic pathway.

248 Despite having similar $\delta^{15}\text{N}$ values, red mullet presented significantly higher THg concentrations than sole. This difference may result from differences in
249 living and feeding habitat. Sole samples were caught in Landes coast, on a sandy-muddy shore, whereas red mullet were caught on rocky shore of the Basque
250 coast. Isotopic difference between the species may support this assumption. Less negative $\delta^{13}\text{C}$ value is commonly interpreted as a higher dependency on
251 benthic-derived organic matter (Carlier et al., 2007; Cresson et al., 2020; Duffill Telsnig et al., 2019; Kopp et al., 2015; Le Loc'h et al., 2008). Isotopic ratios
252 measured for sole can thus track the dependency of this species on the organic matter from the soft bottom sediment, potentially processed by the microbial
253 pathway. On the rocky shore substrate where mullets were caught, pelagic production can enter directly in the food web after falling, since bacterial processes
254 can be of less importance. In addition, mullets were caught close to the mouth of the Nivelle and Bidasoa Rivers. Riverine organic matter classically exhibit
255 very negative carbon isotopic ratios, close to -30‰ (Savoie et al., 2012). Values measured for the Adour river in the present ($-27.84 \pm 0.93\text{‰}$) or previous
256 studies (Bardonnet and Riera, 2005; Liénart et al., 2016) fall within this order of magnitude. In addition, Adour and others Basque rivers are sources of Hg for
257 coastal areas (Sharif et al., 2014) notably since the Adour estuarine area is an area of MeHg production during the dry period by remobilization and
258 methylation of IHg in surface sediment accumulated during the seasonal river inputs (Stoichev et al., 2004). Both isotopic and Hg values may thus testify a
259 higher importance of riverine inputs to red mullet than to sole. But in the absence of accurate $\delta^{15}\text{N}$ values for the actual dietary items of both species (*i.e.*
260 omnivorous crustaceans for red mullet, and suspension feeding bivalves for sole), inferring trophic level of sole and mullet directly from their $\delta^{15}\text{N}$ values
261 should be done cautiously. Species can differ in their actual trophic level while exhibiting similar $\delta^{15}\text{N}$ values (Harmelin-Vivien et al., 2009). In this case,

262 difference in trophic level may originate difference in Hg concentrations. Overall, this study confirms the importance of food sources within the feeding
263 habitat/ecosystem as a major driver of Hg bioaccumulation in marine species muscles (Chouvelon et al., 2018; Cresson et al., 2015b).

264

265 *Differential Hg species distribution within organs*

266 As expected, Hg concentrations were higher in muscle and liver. This organotropism of Hg is consistent with other studies about Hg distributions in organs
267 (Kasper et al., 2009; Polak-Juszczak, 2018). Biochemical characteristics of fish tissues, fish metabolism and Hg chemical behavior are the main drivers of
268 these patterns. High Hg concentrations in muscle is classical and is usually explained by the affinity of Hg for thiols groups in some amino acids and thus
269 proteins. Moreover, muscle is known to have a low MeHg demethylation or elimination rate (Maulvault et al., 2016). Liver is well known for being involved
270 in detoxication processes, explaining why metal concentrations are usually higher in this organ than in others, *i.e.* as in sole in the present study. In addition,
271 hake presented a significant difference of Hg concentrations between muscle and liver. These results suggested different capacities or physiological ability of
272 Hg detoxification by the three fish species. MeHg from the fish body can be directly eliminated or indirectly through the liver by a redistribution of MeHg to
273 other organs such as muscle (Wang et al., 2013). Another detoxication pathway is based on the transformation of MeHg to an inert Hg form by conjugating
274 MeHg to selenium or proteins (*e.g.* glutathione and metallothionein), resulting a less harmful chemical form (Penglase et al., 2014). The lowest %MeHg in
275 sole liver could trace an increase importance of this pathway. Several studies demonstrated that flatfish such as the four-spot megrim *Lepidorhombus boscii*
276 developed increased detoxication abilities, as a response to their chronic exposure to contaminant from diet and sediment (García et al., 2000; Martínez-
277 Gómez et al., 2006). Therefore, differences in Hg distribution between tissues can be attributed to species related to physiological processes especially the
278 detoxification strategies involved in Hg elimination and/or biotransformation (Le Croizier et al., 2018).

279 The gonads are composed mainly by lipoglycophosphoprotein, because teleost oocytes are mainly composed by vitelline proteins. Therefore, the proportion of
280 amino acids in fish egg protein is somehow lower than in muscle (Kasper et al., 2009). However, one interesting fact is that, despite low THg concentration in
281 gonads, hake and red mullet present high %MeHg (74% and 78%, respectively) in this tissue. MeHg is the toxic Hg form which can causes disturbance in fish
282 eggs and larval stages (Latif et al., 2001). This result highlights the potential risk linked to MeHg maternal offloading, that is, the process whereby females
283 transfer a portion of their accumulated contaminants to their offspring during reproduction (Govoni et al., 2017; Lyons and Lowe, 2013; Stefansson et al.,
284 2014). Spawning was thus interpreted as a depuration process in Hg for females, in some freshwater fish species (Donald and Sardella, 2010; Hammerschmidt
285 et al., 1999; Johnston et al., 2001).

290 **Conclusions**

291 In summary, this study has investigated and compared Hg species concentrations in the muscle of three commercial fish species. All individuals except four
292 hakes had THg concentrations in muscle above the food safety thresholds determined by the European Commission regulation. We conclude that the
293 distribution of Hg species (organic and inorganic) is fish species specific. Therefore, differences in MeHg bioaccumulation according to fish species are
294 consequences of their diet, their metabolism (especially MeHg demethylation of processes) and their feeding habitat. Results of the present study confirms the
295 importance of considering the environment in which they reside to investigate Hg bioaccumulation in marine fish. A larger sample size involving larger
296 individuals could be considered, to confirm the results presented above and thus strengthen the European monitoring program.

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302

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