



HAL
open science

Mercury in blue shark (*Prionace glauca*) and shortfin mako (*Isurus oxyrinchus*) from north-eastern Atlantic: Implication for fishery management

Sebastian Biton-Porsmoguer, Daniela Bănaru, Charles F. Boudouresque, Ivan Dekeyser, Marc Bouchoucha, Françoise Marco-Miralles, Benoit Lebreton, Gael Guillou, Mireille Harmelin-Vivien

► To cite this version:

Sebastian Biton-Porsmoguer, Daniela Bănaru, Charles F. Boudouresque, Ivan Dekeyser, Marc Bouchoucha, et al.. Mercury in blue shark (*Prionace glauca*) and shortfin mako (*Isurus oxyrinchus*) from north-eastern Atlantic: Implication for fishery management. *Marine Pollution Bulletin*, 2018, 127, pp.131-138. 10.1016/j.marpolbul.2017.12.006 . hal-01833596

HAL Id: hal-01833596

<https://hal.science/hal-01833596v1>

Submitted on 12 Feb 2019

HAL is a multi-disciplinary open access archive for the deposit and dissemination of scientific research documents, whether they are published or not. The documents may come from teaching and research institutions in France or abroad, or from public or private research centers.

L'archive ouverte pluridisciplinaire **HAL**, est destinée au dépôt et à la diffusion de documents scientifiques de niveau recherche, publiés ou non, émanant des établissements d'enseignement et de recherche français ou étrangers, des laboratoires publics ou privés.

1 **Mercury in blue shark (*Prionace glauca*) and shortfin mako (*Isurus oxyrinchus*) from**
2 **north-eastern Atlantic: implication for fishery management**

3
4 Sebastián Biton-Porsmoguer^{a*}, Daniela Bănaru^a, Charles F. Boudouresque^a, Ivan Dekeyser^a,
5 Marc Bouchoucha^b, Françoise Marco-Miralles^b, Benoît Lebreton^c, Gaël Guillou^c, Mireille
6 Harmelin-Vivien^a

7
8 ^a Aix-Marseille University, CNRS/INSU, Toulon University, IRD, Mediterranean Institute of
9 Oceanography (MIO) UM 110, Campus de Luminy, 13288 Marseille, France

10 ^b Institut Français de Recherche pour l'Exploitation de la Mer (Ifremer), Z.P. Brégaillon, CS
11 20203, 83507 La Seyne-sur-Mer, France

12 ^c Littoral Environnement et Sociétés, UMR 7266 CNRS-Université de La Rochelle, 2 Rue
13 Olympe de Gouges, 17000 La Rochelle, France

14
15 **Short abstract (150 words)**

16
17 Pelagic sharks (blue shark *Prionace glauca* and shortfin mako *Isurus oxyrinchus*) caught by
18 long-line fleets in the NE Atlantic were analysed for total mercury (Hg). Hg concentration in
19 muscle increased with size and weight in both species, but at a higher rate in shortfin mako.
20 Spatial variation was observed only in the blue shark with higher Hg values in the North of the
21 Azorean archipelago. These high-level predators are particularly susceptible to bioaccumulate
22 Hg. However, a significant positive relationship between Hg concentration and trophic level
23 ($\delta^{15}\text{N}$) of individuals was observed only in the shortfin mako. Most sharks landed were juveniles
24 which presented Hg concentration lower than the maximum limit allowed by the European
25 Union for marketing. However, concentrations above this threshold were most recorded in blue
26 sharks larger than 250 cm total length and in shortfin makos larger than 190 cm, raising the
27 question of the commercialization of large-sized individuals.

28
29
30
31

32 Abstract

33

34 Pelagic sharks (blue shark *Prionace glauca* and shortfin mako *Isurus oxyrinchus*) caught by
35 long-line Spanish and Portuguese fleets in the NE Atlantic, were sampled at Vigo fish market
36 (Spain) for total mercury (Hg) analysis. Hg concentration in white muscle increased with size
37 and weight in both species, but at a higher rate in shortfin mako than in the blue shark. No
38 difference was found with sex, year and season. Spatial variation was observed in the blue shark
39 with higher Hg values in the North of the Azorean archipelago, but not in the shortfin mako.
40 These high-level predators are particularly susceptible to bioaccumulate contaminants (Hg) in
41 their tissues (muscle). However, a significant positive relationship between Hg concentration and
42 trophic level ($\delta^{15}\text{N}$) of individuals was observed only in the shortfin mako. Most sharks landed
43 were juveniles which presented Hg concentration lower than the maximum limit allowed by the
44 European Union (1 mg kg^{-1} wet weight) for marketing. However, concentrations above this
45 threshold were most recorded in blue sharks larger than 250 cm total length (TL) and in shortfin
46 makos larger than 190 cm TL, raising the question of the commercialization of large-sized
47 individuals.

48

49 KEYWORDS: Mercury · Pelagic sharks · Blue shark · Shortfin mako · Long-line fishery · NE
50 Atlantic

51

52 1. Introduction

53

54 Long-line Spanish and Portuguese fleets which exploit offshore north-eastern Atlantic waters
55 target pelagic sharks, particularly the blue shark *Prionace glauca* (Linnaeus, 1758) and the
56 shortfin mako *Isurus oxyrinchus*, Rafinesque, 1810, along with the swordfish *Xiphias gladius*
57 Linnaeus, 1758 (Torres et al., 2016). For the past 15 years (2001–2015), the mean landings per
58 year of blue shark represented 2 167 tonnes (63% of long-line landings) and 501 tonnes (14%)
59 for the shortfin mako at the fish market of Vigo in Galicia, Spain (Xunta da Galicia, 2008, pers.
60 comm.; ICCAT, 2015). Sharks are essentially sold for human consumption (meat and fin).

61 The blue shark can reach 380 cm in total length (TL) and could live up to 20 years in the North
62 Atlantic (Skomal and Natanson, 2003). Blue shark females are sexually mature at 220 cm TL (5-

63 6 years) and males at 180 cm TL (4-5 years) (Moreno, 2004; Compagno et al., 2005). The
64 shortfin mako presents a heavier body at similar size than the blue shark, a longer maximum size
65 (440 cm) and a longer life span (30 years max) (Natanson et al., 2006). Median size and age at
66 maturity would be about 280 cm TL and 7-10 years for females, which present a larger size than
67 males, and 200 cm TL, 5-6 years for males (Moreno, 2004; Barreto et al., 2016). However,
68 information on age and growth of both shark species is conflicting and still a matter of debate
69 due to difficult and random sampling of these predators (Skomal and Natanson, 2003; Barreto et
70 al., 2016). These two shark species are highly mobile predators able to migrate over thousands of
71 kilometers in the north Atlantic Ocean (Kohler et al., 2002). Spanish and Portuguese long-liners
72 catch mostly small individuals therefore juveniles represent the major part of shark landings at
73 Vigo fish market for both species (73% of blue sharks and 94% of shortfin makos) (Biton-
74 Porsmoguer, 2015).

75 Sharks position as high-level predators in the marine food web (Ferretti et al., 2010) makes
76 them especially susceptible to contain high concentration of contaminants and particularly
77 mercury (Hg) (Storelli et al., 2002), as Hg is known to bioamplify along food webs, increasing
78 with the trophic level of organisms (Harmelin-Vivien et al., 2009, 2012; Lavoie et al., 2013).
79 Trophic level of organisms is routinely estimated by the nitrogen isotopic ratio ($^{15}\text{N}/^{14}\text{N}$),
80 expressed relative to a standard as $\delta^{15}\text{N}$, which tends to increase with the size of individuals and
81 from prey to predator. $\delta^{15}\text{N}$ is then used to follow the transfer and accumulation of contaminants
82 like Hg or PCB in organisms and food webs (Cabana and Rasmussen, 1994; Booth and Zeller,
83 2005; Cossa et al., 2012). Mercury is a highly toxic trace element present in all compartments of
84 the biosphere. It enters marine food webs from natural and anthropogenic sources (Cossa et al.,
85 2009), and foraging pathway is recognized as being the main Hg contamination way (Mathews
86 and Fisher, 2009). Hg is susceptible to impact aquatic ecosystems including commercial species
87 (McKinley and Johnston, 2010), which could result to adverse health effects on humans like
88 toxic effects on the nervous, digestive, cardiovascular and immune systems, and alterations of
89 fetal neurodevelopment (Castoldi et al., 2003; Diez, 2008). As consumption of marine organisms
90 contributes to most Hg intake in humans, a maximum acceptable level in marine products have
91 been laid down by European Commission regulations and set at 1 mg kg^{-1} wet weight (ww) for
92 high-level pelagic predators (European Commission, 2006: Regulation N^o 1881/2006). Fisheries
93 Department from Galician region is supposed to apply the European regulation and must control

94 sanitary state for all landed sea products (Law 11/2008, December 3rd 2008; Xunta da Galicia,
95 2008). But are the sharks landed and commercialized in Galicia fulfil all these requirements?

96 The main goals of the present study were thus to: (i) measure the total mercury concentration
97 in the muscle of sharks caught in the north-eastern Atlantic Ocean and sold at Vigo fish market,
98 (ii) determine the influence of size, weight, sex, trophic level, zones, season and year on Hg
99 content in these sharks, and (iii) consider the possible implications for the fishery management.

100

101

102 **2. Material and methods**

103

104 *2.1. Sampling and stomach content analysis*

105

106 Sharks were caught by Spanish and Portuguese long-line vessels in the north-eastern Atlantic
107 in five zones (A to F) between the Iberian Peninsula and the Azores archipelago (15°-35°W and
108 30°-45°N), in 2012 and 2013 (Fig. 1). A total of 40 blue shark (*Prionace glauca*) and 48 shortfin
109 mako (*Isurus oxyrinchus*) landed at the fish market of Vigo (Spain) were sampled (Table 1).
110 Blue shark and shortfin mako respectively measured from 74 to 284 cm and from 99 to 219 cm
111 total length (TL). White muscle samples were extracted one centimeter beneath the skin from
112 each individual, put in plastic bags and stored frozen at -20°C. Once at the laboratory, samples
113 were cleaned with distilled water before freeze-drying, grinding and analyzing for total mercury
114 (Hg) and nitrogen stable isotopes ($\delta^{15}\text{N}$). Shark stomachs were extracted and stored at -20°C.
115 After identification and weighing (wet weight, ww) of the prey found in stomach contents, those
116 recently consumed (in good state of conservation) were freeze-dried and analyzed for Hg and
117 $\delta^{15}\text{N}$ in the same way as shark muscle samples. Total wet weights of partially digested prey were
118 reconstructed according to the size of their hard pieces (beaks for cephalopods and otoliths or
119 vertebrae for teleost fish) using pre-established relationships (Biton-Porsmoguer, 2015).
120 Reconstructed weight percentages (% ww) of the main prey types found in stomach contents
121 were then determined for both shark species.

122

123 *2.2. Mercury and stable isotope analyses*

124

125 Total Hg concentrations were determined with a semi-automated atomic absorption
126 spectrophotometer AMA 254 (Altec Ltd, Prague, Czech Republic) with a detection limit of 0.003
127 ng/mg, following the procedure described in Cossa et al. (2009). Hg quantification procedure
128 consisted in three automatic sequences: (1) ashing at 550°C of the freeze-dried sample for Hg
129 volatilisation, (2) evolved elemental Hg amalgamation on a gold trap, (3) atomic absorption
130 spectrophotometric measurement of the Hg collected after heating the gold trap at 800°C. The
131 accuracy of measurement was assessed every ten samples using certified reference materials
132 from the National Research Council of Canada (fish muscle tissues DORM-4). Hg concentration
133 level of samples was initially expressed as Hg dry weight (dw) concentration in fish muscle or
134 prey muscle samples. However, as Hg concentrations are expressed in wet weight (ww) in
135 international and European regulations, dry weight concentrations were converted into wet
136 weight concentrations, considering that dw concentration = 5 ww concentration (Cresson et al.,
137 2014). The relevancy of this factor has been corroborated with the samples of this study
138 (personal data).

139 Stable isotope analyses were performed with a continuous flow mass spectrometer (Delta V
140 Advantage, Thermo Scientific®, Bremen, Germany) coupled to an elemental analyzer (Flash EA
141 1112 Thermo Scientific®, Milan, Italy). N stable isotope ratios were expressed in the standard δ
142 notation: $\delta^{15}\text{N}\text{‰} = [(R_{\text{sample}}/R_{\text{standard}}) - 1] \times 10^3$, where R is the ratio $^{15}\text{N}/^{14}\text{N}$ in sample and
143 standard (atmospheric air for nitrogen). Accuracy of measurement was estimated by replicate
144 measurements of internal standard (acetanilide) and was lower than 0.1‰.

145

146 2.3. Data analysis

147

148 *Quantification of Hg concentrations in sharks' diet:* The diets of the blue shark and shortfin
149 mako in the sampling area have been previously studied (Biton-Porsmoguer et al., 2013, 2015).
150 The blue shark mainly fed on cephalopods, then teleost fish and rarely cetaceans, while the
151 shortfin mako mainly preyed on teleost fish, then cephalopods, and occasionally cetaceans and
152 chelonians. The importance in weight (% dw) of each prey type was determined for each species
153 and Hg concentration of their diet was quantified with the equation as in Harmelin-Vivien et al.
154 (2012):

155

$$C_{\text{diet}(i)} = \sum [C_{\text{prey}(x)} \times W_{\text{prey}(x)(i)}]$$

156 where $C_{\text{diet}(i)}$ is the Hg concentration in the diet of the shark species (i), $C_{\text{prey}(x)}$ is the Hg
157 concentration of the prey (x), $W_{\text{prey}(x)(i)}$ is the percentage in weight of the prey (x) in the (i)th
158 shark species, and Σ the sum of the product for all the (x) prey types. In a similar way, the $\delta^{15}\text{N}$
159 value of the diet of each shark species was determined following the equation:

$$160 \quad \delta^{15}\text{N}_{\text{diet}(i)} = \Sigma[\delta^{15}\text{N}_{\text{prey}(x)} \times W_{\text{prey}(x)(i)}]$$

161 where $\delta^{15}\text{N}_{\text{diet}(i)}$ is the $\delta^{15}\text{N}$ of the diet of the shark species (i), $\delta^{15}\text{N}_{\text{prey}(x)}$ is the $\delta^{15}\text{N}$ of the prey
162 (x), $W_{\text{prey}(x)(i)}$ is the percentage in weight of the prey (x) in the (i)th shark species, and Σ the sum
163 of the product for all the (x) prey types. These calculations represent only estimations as Hg
164 concentrations and $\delta^{15}\text{N}$ are measured on prey muscles, while weights include all prey tissues.

165
166 *Biomagnification factor calculations:* A mean biomagnification factor (BMF) was determined
167 for the two sharks in the north-eastern Atlantic following Fisk et al. (2001), based on mean Hg
168 concentration and trophic level of sharks and their diet, using the following equation:

$$169 \quad \text{BMF}_{\text{shark}(i)} = [(C_{\text{shark}(i)}/C_{\text{diet}(i)})/(\delta^{15}\text{N}_{\text{shark}(i)}/\delta^{15}\text{N}_{\text{diet}(i)})]$$

170 Where $C_{\text{shark}(i)}$ is the Hg concentration in the shark species (i), $C_{\text{diet}(i)}$ is the Hg concentration in
171 the diet of the (i) shark species, $\delta^{15}\text{N}_{\text{shark}(i)}$ is the trophic level of the (i) shark species, and
172 $\delta^{15}\text{N}_{\text{diet}(i)}$ is the trophic level of the diet of the (i) shark species. This factor is based on the
173 assumption that mercury concentration in a predator depends on those of its prey, corrected for
174 trophic level difference between predator and prey. We replace the trophic level estimation used
175 by Fisk et al. (2001) by $\delta^{15}\text{N}$ values, which represents a more accurate estimation of organism
176 trophic position.

177
178 *Statistical analysis:* As the number, size, weight and sex of individuals differed between
179 sampling zones and species, differences were tested by ANCOVA to remove the effects of these
180 parameters and consider only the factor tested. When significant differences were observed
181 Tukey post-hoc tests were performed. Relationships between Hg content and sharks size, weight
182 and $\delta^{15}\text{N}$ were tested by Pearson's linear correlation on \log_{10} transformed Hg concentrations to
183 linearize the regression and stabilize the variances. Differences in slope and elevation were tested
184 by appropriate *t*-test. *T*-test was also used for interspecific comparisons of Hg and $\delta^{15}\text{N}$ values.

185
186

187 3. Results

188

189 3.1. Influence of biological parameters on Hg level

190

191 In both species, mercury concentration did not vary with sex for individuals of similar size or
192 weight (ANCOVA, all $p > 0.05$). Thus, sexes were combined for further analyses. Total Hg level
193 in muscle ranged from 0.14 to 1.71 mg kg⁻¹ ww in the blue shark and from 0.12 to 2.57 mg kg⁻¹
194 ww in the shortfin mako (Table 2). Mean Hg concentration and mean $\delta^{15}\text{N}$ value were
195 significantly higher in the shortfin mako than in the blue shark (t -tests, $t = -2.213$, $p = 0.029$ and t
196 $= -2.848$, $p = 0.006$ respectively) (Table 2). Hg concentration increased significantly with size
197 and weight in both species, as significant linear relationships were observed between log Hg and
198 TL, (cm) and between log Hg and total weight (W kg ww) (log Hg = 0.009 W - 0.555, $R^2 = 0.59$,
199 $p < 0.001$ in blue shark; log Hg = 0.012 W - 0.630, $R^2 = 0.66$, $p < 0.001$ in shortfin mako).
200 Whatever the parameter considered (size or weight), the slope of the regression was significantly
201 higher (t -test, $p < 0.05$) for the shortfin mako than the blue shark, suggesting different
202 bioaccumulation capacities between the two species. However, the relationship between Hg
203 concentration and the trophic level of individual, expressed as $\delta^{15}\text{N}$ values, was significant only
204 for the shortfin mako (log Hg = 0.169 $\delta^{15}\text{N} - 2.181$, $R^2 = 0.10$, $p = 0.036$), but not for the blue
205 shark (log Hg = -0.069 $\delta^{15}\text{N} + 0.422$, $R^2 = 0.02$, $p = 0.197$).

206

207 3.2. Geographical and temporal variations

208 In blue sharks of similar size, significantly higher mean Hg concentrations (x1.6) were
209 recorded in individuals collected in zone A, than in zone B, D and F, in which blue shark had
210 similar lower concentrations (ANCOVA $F = 3.46$, $p = 0.026$) and weight (ANCOVA $F = 3.37$, p
211 $= 0.029$). Pearson's linear correlations between log Hg and size presented similar slopes in all
212 zones, but a higher intercept value in zone A (Fig. 3), suggesting the influence of environmental
213 differences between zone A in the North of the Azores archipelago and the other zones, rather
214 than a biological difference between individuals. In contrast, no difference of Hg level among
215 zones was observed for the shortfin mako, taken into account differences in size or weight of
216 individuals in the different zones (ANCOVA, $F = 2.46$, $p = 0.086$ for size; $F = 1.49$, $p = 0.242$
217 for weight). In both species and all zones combined, Hg concentration in muscle did not

218 significantly vary with year (2012 vs 2013) nor season (spring vs autumn) (ANCOVA, all $p >$
219 0.05).

220

221 3.3. Biomagnification factor and influence of diet

222

223 The biomagnification factor BMF takes only into account the last trophic level of the food web
224 analyzed (the predator and its prey). Hg level and $\delta^{15}\text{N}$ of the main prey ingested by these two
225 sharks displayed highly variable inter- and intraspecific values (Table 3). The lowest mean Hg
226 concentrations were recorded in pelagic teleost prey fish, such as *Scomber* sp. ($0.02 \text{ mg kg}^{-1} \text{ ww}$)
227 and *Scomberesox saurus* ($0.08 \text{ mg kg}^{-1} \text{ ww}$), and the highest ones in some cephalopods (*Illex* sp.,
228 $2.24 \text{ mg kg}^{-1} \text{ ww}$) and the cetacean *Delphinus delphis* ($1.77 \text{ mg kg}^{-1} \text{ ww}$). The lowest $\delta^{15}\text{N}$ value
229 was observed in the teleost fish *Euthynnus alletteratus* (9.7‰) and the highest one in the
230 cephalopod *Ancistroteuthis lichtensteinii* (12.9‰). No significant correlation was found between
231 log Hg and $\delta^{15}\text{N}$ in prey, or $\delta^{15}\text{N}$ in prey and sharks combined ($R^2 = 0.06$, $p > 0.05$ for both
232 regressions). The blue shark and shortfin mako ingested then prey with various Hg burden, some
233 presenting higher Hg concentration and trophic level than them. The blue shark mainly fed on
234 cephalopods (76% by weight), teleost fish (18%) and cetaceans (0.03%), while the shortfin mako
235 preyed mainly on teleost fish (66% by weight), cephalopods (27%), cetaceans (0.05%) and sea
236 turtles (0.03%). When growing, the blue shark consumed less cephalopods and more teleosts and
237 cetaceans, while the shortfin mako ingest less teleosts and more cetaceans and chelonians. Mean
238 Hg concentration and $\delta^{15}\text{N}$ in diet was higher for the blue shark than for shortfin mako, globally
239 and in most size classes (Table 4), while the reverse was observed in shark muscle with generally
240 higher Hg level in shortfin mako muscle than in blue shark muscle. Mean BMF value was low
241 (<1) in the blue shark, but higher than 1 in the shortfin mako (Table 4), suggesting Hg
242 bioamplification from diet to predator in the shortfin mako only. When the different size classes
243 were considered, it was observed that BMF tended to increase with the size of individuals in both
244 species, but not in a linear way. BMF remained always <1 in the blue shark suggesting no Hg
245 bioamplification in this species. In the shortfin mako, no bioamplification was recorded in the
246 smaller individuals ($<130 \text{ cm TL}$) with $\text{BMF} <1$, while Hg bioamplification occurred for larger-
247 sized individuals (particularly $>200 \text{ cm TL}$) with BMF largely >1 . Such results indicated no

248 straightforward relationship between Hg level of prey and predator, but rather suggested
249 differences in metabolism and/or prey consumption rate between the two shark species.

250

251 3.4. *Mercury in sharks and food safety*

252

253 The significant exponential relationship evidenced between Hg level in muscle and body size
254 of sharks revealed that some individuals presented higher mean Hg concentration than the
255 European regulatory threshold ($1.0 \text{ mg kg}^{-1} \text{ ww}$) for the commercialization of high-level pelagic
256 predators. Hg level above this value was observed for 4 individuals over 200 cm TL in the blue
257 shark and for 12 individuals over 150 cm TL in the shortfin mako (Fig 4 and 5). We defined (i) a
258 *size range of potential risk* for consumers with individuals between 200-250 cm TL for the blue
259 shark (Fig 4), and 150-190 cm TL for shortfin mako (Fig 5), as such highly contaminated sharks
260 were present but not numerous in this size range, and (ii) a *size at risk* (250 cm TL for the blue
261 shark and 190 cm TL for the shortfin mako) above which most individuals presented higher Hg
262 level than the allowed EU limit.

263

264 4. Discussion

265

266 4.1. *Bioaccumulation of Hg in blue shark and shortfin mako*

267

268 In both shark species Hg concentration in muscle was positively correlated with size and thus
269 age (Fig. 2), independently of sex, suggesting similar feeding habits in males and females, as
270 observed by Biton-Porsmoguer et al. (2016) with stomach content analysis. Increase of Hg level
271 in organism as they were growing and getting older is a well-known bioaccumulation process in
272 marine organisms, particularly in teleost fish (Cossa et al., 2012; Cresson et al., 2014) and high-
273 level predators like sharks (Storelli et al., 2001, 2002; Branco et al., 2004, 2007; Torres et al.,
274 2017) and cetaceans (André et al., 1991). This age-related increase in Hg content is related to an
275 efficient accumulation of mercury due to the decrease of both detoxification capability and
276 growth rate (Amlund et al., 2007; Dang and Wang, 2012). But many parameters can influence
277 the bioaccumulation of Hg in organism along with age, such as growth rate, reproduction,
278 metabolic activity, Hg concentration in prey and feeding rate (Trudel and Rasmussen, 2006;

279 Cossa et al., 2012). Diet is recognized as the main pathway of Hg intake in high-level predators
280 (Boening, 2000). Both shark species ingested prey of various Hg content and $\delta^{15}\text{N}$ values, some
281 being more contaminated and positioned at a higher trophic level than sharks (Table 3). Hg
282 concentration and $\delta^{15}\text{N}$ in diet did not increase with shark size (Table 4). Thus, the hypothesis of
283 an increase in Hg uptake when sharks were growing linked to an increase in Hg content of diet
284 was not supported by our study. Larger sharks did not always seem to feed at a higher more
285 contaminated trophic level than smaller ones, as observed by McMeans et al. (2010) in Arctic
286 sharks. Feeding on larger prey did not imply to feed on more contaminated or higher trophic
287 level organisms. No relationship between Hg concentration and $\delta^{15}\text{N}$ was found in the blue
288 shark, as also observed by Torres et al. (2017) south of the Azores and Escobar-Sánchez et al.
289 (2010) at Baja California, or by Rumbold et al. (2014) for coastal sharks in Florida. Conversely,
290 positive relationship between Hg and $\delta^{15}\text{N}$ was observed in shortfin mako. This result was also
291 observed in a few shark species analyzed in Australia (Pethybridge et al., 2011) or in the Celtic
292 sea (Domi et al. 2005). Hg is efficiently accumulated in shark tissues with a very slow
293 elimination rate as in teleost fish (Amlund et al., 2007). In addition, the fractionation and turn-
294 over of $\delta^{15}\text{N}$ between diet and shark tissue seems to be also slower in sharks than teleost fish
295 (Hussey et al. 2012), suggesting particular metabolic activity related to nitrogen cycling in
296 elasmobranchs. Torres et al. (2017) recorded also similar Hg concentrations and $\delta^{15}\text{N}$ values in
297 male and female blue sharks. The absence of influence of sex, season and year on mercury level
298 in both species can be explained by the high mobility of these sharks regardless of sex (Queiroz
299 et al., 2016) and the large spectrum of prey consumed at different seasons during the two years
300 (Biton-Porsmoguer et al., 2013, 2015).

301 A higher increase in Hg content with size was observed in the shortfin mako compared with
302 blue shark (Fig. 2), suggesting different bioaccumulation processes between the two species
303 (age, diet, activity and/or metabolism). While the blue shark and the shortfin mako are long-lived
304 species (with a higher life span for shortfin mako) (Skomal and Natanson, 2003; Natanson et al.,
305 2006), the age of specimens analyzed in this study ranged likely from 1 to 10 years for both
306 species with a majority of juveniles (87.5%), especially in the shortfin mako (Biton-Porsmoguer,
307 2015). Shortfin makos were heavier than blue sharks at the same size, invalidating the possibility
308 of a dilution of Hg by growth in the blue shark to explain its lower Hg content. Higher Hg
309 content of diet in shortfin mako was also not supported by our results (Table 4). Thus, the higher

310 Hg concentration observed in the shortfin mako could not be attributed to an older age, a slower
311 growth rate or a higher Hg content of its prey. Differences in metabolic activity and feeding rate
312 could explain the higher Hg uptake with size observed in shortfin mako compared to blue shark.
313 Shortfin mako belongs to the warm-bodied Lamnidae which present an elevated aerobic
314 metabolism compared to their ectothermic relatives like the blue shark which belongs to the
315 Carcharhinidae (Shadwick and Goldbogen, 2012). The shortfin mako can maintain higher
316 musculature, brain, eyes and viscera temperatures than the surrounding water (1 to 10°C higher
317 than ambient temperature) (Carey, 1982). Therefore, it needs to eat more frequently than the blue
318 shark and accumulates more mercury due to an increased feeding rate. This hypothesis was
319 supported by the lower vacuity index (percentage with an empty stomach) observed in the
320 shortfin mako (36%) compared to blue shark (47%) (Biton-Porsmoguer, 2005).

321

322 *4.2. Regional differences*

323

324 Mercury levels recorded in the blue shark in this study (0.14-1.71 mg kg⁻¹ ww) were in
325 agreement with values previously reported in different studies performed in the north Atlantic
326 Ocean (0.16-1.84 mg kg⁻¹ ww) and Tasmania (SW Pacific Ocean) (0.27-1.20 mg kg⁻¹ ww), but
327 not on the south coast of Brazil (0.46-2.40 mg kg⁻¹ ww) (De Carvalho et al., 2014) where higher
328 Hg concentrations were recorded (Table 5). This may be related to the high mobility of these
329 sharks, their ability to cover large areas for feeding and their dietary opportunism. However,
330 Branco et al. (2007) record a higher accumulation rate of total mercury with size in the blue
331 shark from the equatorial Atlantic (0.68-2.50 mg kg⁻¹ ww) compared with specimens from the
332 Azores archipelago (0.22-1.30 mg kg⁻¹ ww). They relate this pattern to higher Hg concentration
333 in prey, as well as differences in quantity and type of food eaten in the Equator. Such a
334 hypothesis was not relevant in our study, as the rate (regression slope) of Hg increase in blue
335 shark with size was similar in all zones (Fig. 3). Higher Hg concentrations in the north of Azores
336 could not be attributed to gender, as no difference with sex was observed. In mid-Atlantic (south
337 of the Azores), Torres et al. (2017) record lower Hg concentrations in the blue shark than we did
338 for individuals of similar size range in the same area (0.33 ± 0.02 and 0.41 ± 0.02 respectively).
339 The higher intercept value of the linear regression for specimens from the north of the Azores
340 suggested rather the influence of environmental conditions with probably higher Hg

341 concentrations in sea water and prey in this zone, but no difference in metabolism, feeding rate
342 or prey types between populations. The Azores archipelago is a volcanic region with naturally
343 high mercury level in sea water and sediment (Guest et al., 1999) that may explain the
344 significantly higher levels of Hg found in the blue sharks of zone A. Moreover, the Azores
345 archipelago constitutes probably a nursery area for the blue shark and juveniles can stay in this
346 area at least for two years (Vandeperre et al., 2014), accumulating thus a higher Hg burden
347 during this time. However, in the absence of Hg concentration analysis in seawater and in a
348 higher number of prey in the different zones, we cannot test this hypothesis.

349 No difference in Hg concentration among zones was observed for the shortfin mako in this
350 study. This homogeneity was probably due to the high swimming activity of this species, which
351 moves frequently between distant zones (Kohler et al., 2002). The Hg values recorded in the
352 shortfin mako were high (0.12-2.57 mg kg⁻¹ ww) and in the range of those observed for this
353 species in the north Atlantic and other oceans (0.15-3.12 mg kg⁻¹ ww) (Table 5).

354

355 *4.3. Biomagnification of Hg in sharks*

356

357 Mercury, under its organic monomethylmercury form, is one of the few trace metals that
358 biomagnifies along food webs (Gray, 2002). Biomagnification is assumed when BMF factor
359 (concentration in predator/concentration in diet corrected from their respective trophic level) is
360 higher than 1 (Fisk et al., 2001). Biomagnification of Hg was observed only in the shortfin mako,
361 not in the blue shark (Table 4). These two sharks consumed prey with a great variability in both
362 Hg concentration and $\delta^{15}\text{N}$ value (Table 3). For the blue shark, Hg content in diet was higher
363 than in shark muscle for all size classes, leading up to a BMF always <1, while slightly
364 increasing with size. For the shortfin mako, higher Hg content in shark muscle compared with
365 diet and BMF values >1 were observed from individuals larger than 130 cm, and more
366 significantly larger than 200 cm. However, BMF calculation should be considered cautiously.
367 First, the reduced number of prey analysed in this study is not sufficient to characterize the
368 global diet of the two species and a broader study should be conducted to conclude. Second, the
369 same mean Hg and $\delta^{15}\text{N}$ values of prey groups were used for all size-classes of both sharks.
370 However, as observed by Biton-Porsmoguer et al. (2015), they could prey on different species.
371 Finally, the time lag between ingestion and assimilation of prey was not considered. Stomach

372 content offered a snap shot of shark's diet when caught, while muscle Hg concentration reflected
373 food assimilation over several months. Nonetheless, similar results are reported by Maz-Courrau
374 et al. (2012) at Baja California with Hg biomagnification in the shortfin mako but not in the blue
375 shark. The absence of any Hg biomagnification in blue shark is also observed in the Pacific
376 Ocean of México by Escobar-Sánchez et al. (2011). Difference in BMF factor between the two
377 species, with Hg biomagnification occurring only in the shortfin mako is probably related with
378 their metabolic differences (endothermy in shortfin mako vs ectothermy in blue shark)
379 (Shadwick and Goldbogen, 2012) as discussed above, with a positive relationship between Hg
380 content and $\delta^{15}\text{N}$ in the shortfin mako and not in blue shark.

381

382 4.4. Food safety and fishery implication

383

384 The mean values of mercury level in the muscle of blue shark and shortfin mako analyzed in
385 this study were lower than the upper limit allowed by the European Union in high-level pelagic
386 predators for human consumption ($1 \text{ mg kg}^{-1} \text{ ww}$) (Table 2), as also observed by Torres et al.
387 (2017) for these species. However, Hg concentration exceeded the legal EU standard in larger
388 individuals (Fig. 4 and 5). Above 250 cm TL in *P. glauca* and 190 cm TL in *I. oxyrinchus* most,
389 if not all, individuals exhibited Hg content well above the EU sanitary risk limit, representing a
390 *size at risk* for the consumption of these species. Measuring mercury concentration above
391 European and international regulatory thresholds in shark muscle is regularly reported in the
392 literature, whatever the ecology, geographic distribution or feeding habits of species (Storelli et
393 al., 2002; Branco et al., 2004, 2007; Pethybridge et al., 2010; Escobar-Sánchez et al., 2011; De
394 Carvalho et al., 2014; Cresson et al., 2014; Mc Kinney et al., 2016, and references in Table 5).
395 All authors raise the health issue of shark, and more generally high-level predator, consumption.
396 However, Hg concentration value alone does not seem to be sufficient to evaluate the toxicity of
397 marine organisms for human consumption, but rather the molar ratio Se:Hg, as selenium is
398 known to play a protective role against the toxic effects of Hg and enhance detoxifying
399 mechanisms (Branco et al., 2007; Khan and Wang, 2009). But studies on sharks record low
400 Se:Hg values (Kaneko and Ralston, 2007; Escobar-Sánchez et al., 2011) or no relationship
401 between Se and Hg concentrations (Torres et al., 2017), which leaves unsold the problem of Hg
402 toxicity in larger sharks.

403 The commercialisation of blue shark (>250 cm TL) and shortfin mako (>190 cm TL) exceeding
404 the *size at risk* is thus an issue for Galician Fishery Authorities in terms of compliance with
405 European legislation. The shark fisheries are not regulated (no quotas and size limits) in Spain.
406 According to our results, in terms of human food safety, we wonder about the possible risk
407 linked to large shark consumption, especially when they are caught between the Azores
408 Archipelago and the Iberian Peninsula. However, limiting large shark consumption may lead to
409 an increase in finning practice (prohibited in the EU in 2003 with application for Spain and
410 Portugal in June 2013). Therefore, maritime control should be enforced for the strict application
411 of the Council Regulation (EC No 1185/2003). Furthermore, some nursery areas, such as the
412 Azores Archipelago (Vandeperre et al., 2014), Gulf of Cadix (Compagno et al., 2005), and north
413 of Galicia (Biton-Porsmoguer, pers. com.), may concentrate big blue shark females exceeding
414 *the size at risk* for mercury level. Within these areas shark fishing should be limited or at least
415 regulated. No information about reproduction activities has been recorded for shortfin mako up
416 to now.

417

418 **5. Conclusions**

419

420 Mercury concentrations in muscle of the blue shark *P. glauca* and the shortfin mako *I.*
421 *oxyrinchus* in north-eastern Atlantic were in the range of values reported in other regions for
422 these widely-distributed sharks. An increase in Hg burden with length and weight was observed
423 in both species, but at a higher rate in the shortfin mako. Bioaccumulation of Hg with individual
424 trophic level ($\delta^{15}\text{N}$) and biomagnification from prey to predator was only observed in the shortfin
425 mako, and likely related to its endothermic metabolism and higher feeding activity. The absence
426 of significant difference between sexes, seasons and years in both species could be explained by
427 the high mobility of these sharks and their feeding adaptation to different environmental
428 conditions. Most blue shark and shortfin mako landed and sold at the fish market of Vigo (Spain)
429 exhibited Hg concentration in muscle lower than the maximum limit allowed by the European
430 Union for human consumption ($1 \text{ mg kg}^{-1} \text{ ww}$), as they were juveniles. However, Hg content
431 above this legal threshold was recorded in the majority of adult blue shark larger than 250 cm TL
432 and shortfin mako larger than 190 cm TL. Then, we recommend avoiding the capture and
433 commercialization of individuals exceeding these respective lengths for the two species. In

434 addition, a size range of potential risk was defined (from 200 to 250 cm for the blue shark and
435 from 150 to 190 cm for the shortfin mako) in which some individuals might present Hg
436 concentration above the legal limit. The landing of blue shark and shortfin mako in Vigo during
437 the next decade should assume the implementation of management measures for the sustainable
438 fishing exploitation and the conservation of these species.

439

440 **Acknowledgments**

441

442 We gratefully acknowledge the Port Authority of Vigo (Spain), which authorized sampling, and
443 the Captains of the fishing boats for allowing us to measure and dissect landed sharks before
444 selling them. Mercury analyses were realized by Ifremer Laboratory at La Seyne-sur-Mer
445 (France). Stable isotope analyses were performed at the LIENSs laboratory, University of La
446 Rochelle (France). Thank are also due to Michael Paul, a native English speaker, for English
447 correction, and to an anonymous reviewer for most helpful comments.

448

449 **References**

- 450 Amlund, H., Lundebye, A.K., Berntssen, M.H.G., 2007. Accumulation and elimination of
451 methylmercury in Atlantic cod (*Gadus morhua* L.) following dietary exposure. *Aquat.*
452 *Toxicol.* 83, 323–330.
- 453 André, J., Boudou, A., Ribeyre, F., Bernhard, M., 1991. Comparative study of mercury
454 accumulation in Dolphins (*Stenella coeruleoalba*) from French Atlantic and
455 Mediterranean coasts. *Sci. Total Environ.* 104, 191–209.
- 456 Barreto, R.R., de Farias, W.K.T, Andrade, H., Santana, F.M., Lessa R., 2016. Age, growth and
457 spatial distribution of the life stages of the shortfin mako, *Isurus oxyrinchus* (Rafinesque,
458 1810) caught in the western and central Atlantic. *PLoS One.* 11(4), e0153062.
- 459 Biton-Porsmoguer, S., 2015. Biologie, écologie et conservation du requin peau bleue (*Prionace*
460 *glauca*) et du requin mako (*Isurus oxyrinchus*) en Atlantique nord-est. Thèse doctorale.
461 Aix-Marseille Université. 269p.
- 462 Biton-Porsmoguer, S., Bănaru, D., Béarez, P., Dekeyser, I., Merchán Fornelino, M., Míguez-
463 Lozano, R., Boudouresque, C-F., 2013. Diet of the shortfin Mako *Isurus oxyrinchus* and

464 the blue shark *Prionace glauca* in the northeastern Atlantic Ocean. Rapp. Comm. int.
465 Mer Médit. 40, 499.

466 Biton-Porsmoguer, S., Bănaru, D., Boudouresque, C-F., Dekeyser, I, Merchán, M., 2015. DNA
467 evidence of the consumption of short-beaked common dolphin *Delphinus delphis* by the
468 shortfin mako *Isurus oxyrinchus*, in the Northeastern Atlantic Ocean, Mar. Ecol. Progr.
469 Ser. 532, 177–183.

470 Biton-Porsmoguer, S., Dekeyser, I., Bănaru, D., Bouchoucha, M., Marco-Miralles, F., 2016.
471 Using the body mass index (BMI) to estimate mercury contamination of the blue shark
472 (*Prionace glauca*) and the shortfin mako (*Isurus oxyrinchus*). Cybium 40(4), 319–322.

473 Boening, D.X., 2000. Ecological effects, transport, and fate of mercury: a general review.
474 Chemosphere 40, 1335–1351.

475 Booth S., Zeller D., 2005. Mercury, food webs, and marine mammals: implications of diet and
476 climate change for human health. Environ. Health Persp. 113, 521–526.

477 Branco, V., Canário, J., Vale, C., Raimundo, J., Reis, C., 2004. Total and organic mercury
478 concentrations in muscle tissue of the blue shark (*Prionace glauca* L.1758) from the
479 Northeast Atlantic. Baseline / Mar. Pollut. Bull. 49, 854–874.

480 Branco, V., Vale, C., Canário, J., Santos, M.N., 2007. Mercury and selenium in blue shark
481 (*Prionace glauca*, L. 1758) and swordfish (*Xiphias gladius*, L. 1758) from two areas of
482 the Atlantic Ocean. Environ. Pollut. 150, 373–380.

483 Cabana, G., Rasmussen, J.B., 1994. Modelling food chain structure and contaminant
484 bioaccumulation using stable nitrogen isotopes. Nature 372, 255–257.

485 Carey, F.G. 1982. Warm fish. In: C. R. Taylor, K. Johansen & L. Bolis, eds., A companion to
486 animal physiology. Cambridge University Press, Chapt. 16, 216–233.

487 Castalodi, A.F., Coccini, T., Manzon, L., 2003. Neurotoxic and molecular effects of
488 methylmercury in humans. Rev. Environ Health. 18, 19–31.

489 Compagno, L., Dando, M., Fowler, S., 2005. Guía de campos de los tiburones del mundo.
490 Barcelona, Ediciones Omega, 357p.

491 Cossa, D., Averty, B., Pirrone, N., 2009. The origin of methylmercury in open Mediterranean
492 waters. Limnol. Oceanogr. 54, 837–844.

493 Cossa, D., Harmelin-Vivien, M., Mellon-Duval, L., Loizeau, V., Averty, B., Crochet, S., Chou,
494 L., Cadiou, J.-F. 2012. Influence of bioavailability, trophic position and growth on

495 methylmercury in hakes (*Merluccius merluccius*) from northwestern Mediterranean and
496 northeast Atlantic. Environ. Sci. Technol. 46, 4885–4893.

497 Council Regulation (EC) N°. 1185/2003 of 26 June of 2003 on the removal of fins of sharks on
498 board vessels. Official Journal of the European Union L 167/1, 26 June 2003, 3p.

499 Cresson, P., Fabri, M.C., Bouchoucha, M., Brach Papa, C., Chavanon, F., Jadaud, A., Knoery, J.,
500 Miralles, F., Cossa, D. 2014. Mercury in organisms from the Northwestern
501 Mediterranean slope: Importance of food sources. Sci. Total Environ. 497–498, 229–238.

502 Dang, F., Wang, W.X., 2012. Why mercury concentration increases with fish size? Biokinetic
503 explanation. Environ. Pollut. 163, 192-198.

504 Davenport, S., 1995. Mercury in blue sharks and deepwater dogfish from around Tasmania.
505 Australian Fish. 54 (3), 20–22.

506 De Carvalho, G.G.A., Degaspari, I.A.M., Branco, V., Canário, J., Ferreira de Amorim, A.,
507 Kennedy, V.H., Ferreira, J.R., 2014. Assessment of total and organic mercury levels in
508 blue sharks (*Prionace glauca*) from the South and Southeastern Brazilian Coast. Trace
509 Elem. Res. 159, 128–134.

510 Dias, A.C.L., Guimarães, J.R.D., Malm. O., Costa, P.A.S., 2008. Total mercury in muscle of the
511 shark *Prionace glauca* (Linnaeus, 1758) and swordfish *Xiphias gladius* Linnaeus, 1758,
512 from the South-Southeast coast of Brazil and the implications for public health. Cad.
513 Saúde Pública, Rio de Janeiro. 24(9), 2063–2070.

514 Díez, S., 2008. Human health effects of methylmercury exposure. In: D.M. Whitacre (ed.) Rev.
515 Environ. Contam. T. 198, 111–132.

516 Domi N., Bouquegneau J.M., Das K., 2005. Feeding ecology of five commercial shark species
517 from the Celtic Sea through stable isotope and trace metal analysis. Mar. Environ. Res.
518 60(5), 551–569.

519 Escobar-Sánchez, O., Galván-Magaña, F., Rosíles-Martínez, R., 2011. Biomagnification of
520 mercury and selenium in blue shark *Prionace glauca* from the Pacific Ocean of Mexico.
521 Biol. Trace Elem. Res. 144, 550–559.

522 European Commission., 2006. Commission Regulation (EC) No 1881/2006 of 19 December
523 2006 setting maximum levels for certain contaminants in foodstuffs. Off. J. Eur.
524 Communities: Legis 37:4. ELI: <http://data.europa.eu/eli/reg/2006/1881/2015-05-21>.

525 Ferretti, F., Worm, B., Britten, G.L., Heithaus, M.R., Lotze, H.K. 2010. Patterns and ecosystem
526 consequences of shark declines in the ocean. *Ecol. Lett.* 13, 1055–1071.

527 Fisk, A.T., Hobson, K.A., Norstrom, R.J., 2001. Influence of chemical and biological factors on
528 trophic transfer of persistent organic pollutants in the Northwater Polynia marine food
529 web. *Environ. Sci. Technol.* 35, 732–738.

530 Gray, J.S., 2002. Biomagnification in marine systems: the perspective of an ecologist. *Mar. Poll.*
531 *Bull.* 45, 46–52.

532 Guest, J. E., Gaspar, J. L., Cole, P. D., Queiroz, G., Duncan, A. M., Wallestein, N., Ferreira, T.,
533 Pacheco, J-M., 1999. Volcanic geology of Furnas Volcano, Sao Miguel, Azores. *J.*
534 *Volcanol.* 92, 1–29

535 Harmelin-Vivien, M., Bodiguel, X., Charmasson, S., Loizeau, V., Mellon-Duval, C.,
536 Tronczyński, J., Cossa, D., 2012. Differential biomagnification of PCB, PBDE, Hg and
537 Radiocesium in the food web of the European hake from the NW Mediterranean. *Mar.*
538 *Poll. Bull.* 64, 974–983.

539 Harmelin-Vivien, M., Cossa, D., Crochet, S., Bănar, D., Letourneur, Y., Mellon-Duval, C.,
540 2009. Difference of mercury bioaccumulation in red mullets from the north-western
541 Mediterranean and Black seas. *Mar. Poll. Bull.* 58, 679–685.

542 Hussey, N.E., MacNeil, M.A., Olin, J.A., McMeans, B.C., Kinney, M.J., Chapman, D.D., Fisk,
543 A.T., 2012. Stable isotopes and elasmobranchs: tissue types, methods, applications and
544 assumptions. *J. Fish Biol.* 80, 1449–1484.

545 ICCAT., 2015. Report of the 2015 ICCAT Blue Shark stock assessment session. Portugal 2015,
546 1–115.

547 Instituto Español de Oceanografía (IEO)., 2003. Niveles de arsénico y metales pesados en
548 Marrajos (*Isurus oxyrinchus*), Pez espada (*Xiphias gladius*) y Tintorera (*Prionace*
549 *glauca*). Informe final. Convenio específico de colaboración entre la Secretaría General
550 de Pesca Marítima y el Instituto Español de Oceanografía. 48p.

551 Kaneko, J.J., Ralston, N.V., 2007. Selenium and mercury in pelagic fish in the central North
552 Pacific near Hawaii. *Biol. Trace Elem. Res.* 119, 242–254.

553 Khan, M.A.K., Wang, F., 2009. Mercury-selenium compounds and their toxicological
554 significance: Toward a molecular understanding of the mercury-selenium antagonism.
555 *Environ. Toxicol. Chem.* 28, 1567–1577.

556 Kohler, N.E., Turner, P.A., Hoey, J.J., Natanson L.J., Briggs R., 2002. Tag and recapture data for
557 three pelagic shark species: blue shark (*Prionace glauca*), shortfin mako (*Isurus*
558 *oxyrinchus*) and porbeagle (*Lamna nasus*) in the north Atlantic Ocean. Coll. Vol. Sci.
559 Pap. ICCAT. 54 (4), 1231–1260.

560 Lavoie, R.A., Jardine, T.D., Chumchal, M.M., Kidd, K.A, Campbell, L.M., 2013.
561 Biomagnification of mercury in aquatic food webs: A worldwide meta-analysis. Environ.
562 Sci. Technol. 47, 13385–13394.

563 Mathews, T., Fisher, N.S., 2009. Dominance of dietary intake of metals in marine elasmobranch
564 and teleost fish. Sci. Total Environ. 407, 5156–5161.

565 Maz-Courrau, A., López-Vera, C., Galván-Magaña, F., Escobar-Sánchez, O., Rosiles-Martínez,
566 R., Sanjuán-Muñoz, A., 2012. Bioaccumulation and biomagnification of total mercury in
567 four exploited shark species in the Baja California Peninsula, Mexico. Bull. Environ.
568 Contam. Toxicol. 88, 129–134.

569 McKinley, A., Johnston, E.L., 2010. Impacts of contaminant sources on marine fish abundance
570 and species richness: a review and meta-analysis of evidence from the field. Mar. Ecol.
571 Progr. Ser. 420, 175–191.

572 McKinney, M.A., Dean, K., Hussey, N.E., Cliff, G., Wintner, S.P., Dudley, S.F.J., Zungu, M.P.,
573 Fisk, A.T., 2016. Global versus local causes and health implications of high mercury
574 concentrations in sharks from the east coast of South Africa. Sci. Total Environ. 541,
575 176–183.

576 McMeans, B.C., Svavarsson, J., Dennard, S., Fisk, A.T., 2010. Diet and resource use among
577 Greenland sharks (*Somniosus microcephalus*) and teleosts sampled in Icelandic waters,
578 using $\delta^{13}\text{C}$, $\delta^{15}\text{N}$, and mercury. Can J Fish Aquat Sci. 67, 1428–1438.

579 Moreno, J.A., 2004. Guía de los tiburones de aguas ibéricas, Atlántico nororiental y
580 Mediterráneo. Barcelona, Omega publ. 315p.

581 Nakano, H. and Stevens, J.D., 2008. The biology and ecology of the blue shark, *Prionace*
582 *glauca*; pp. 140–151. In Sharks of the open ocean: Biology, fisheries and conservation.
583 Edited by M. D. Camhi, E. K. Pikitch, and E. A. Babcock. Blackwell Publishing. Oxford,
584 U.K.

585 Natanson, L.J., Kohler, N.E., Ardizzone, D., Cailliet, G.M., Wintner, S.P., Mollet, H.F., 2006.
586 Validated age and growth estimates for the shortfin mako, *Isurus oxyrinchus*, in the North
587 Atlantic Ocean. *Environ. Biol. Fish.* 77, 367–383.

588 Pethybridge, H., Cossa, D., Butler, E.C.V., 2010. Mercury in 16 demersal sharks from southeast
589 Australia: Biotic and abiotic sources of variation and consumer health implications. *Mar.*
590 *Environ. Res.* 69, 18–26.

591 Queiroz, N., Humphries, N.E., Mucientes, G., Hammerschlag, N., Lima, F.P., Scales, K.L.,
592 Miller, P.I., Sousa, L.L., Seabra, R., Sim, D.W., 2016. Ocean-wide tracking of pelagic
593 sharks reveals extent of overlap with longline fishing hotspots. *Proc. Natl. Acad. Sci.*
594 *U.S.A.* 113(6), 1582–1587.

595 Rumbold, D., Wasno, R., Hammerschlag, N., Volety, A., 2014. Mercury accumulation in sharks
596 from the coastal waters of southwest Florida. *Arch. Environ. Contam. Toxicol.* 67,
597 402–412.

598 Shadwick, R.E., Goldbogen, J.A., 2012. Muscle function and swimming in sharks. *J. Fish. Biol.*
599 80, 1904–1939.

600 Skomal, G.B., Natanson, L.J., 2003. Age and growth of the blue shark (*Prionace glauca*) in the
601 North Atlantic Ocean. *Fish. Bull.* 101, 627–639.

602 Storelli, M.M., Giacomini-Stuffler, R., Marcotrigiano, G.O., 2001. Total mercury and
603 methylmercury in tuna fish and sharks from the south Adriatic Sea. *Ital. J. Food Sci.* 13,
604 101–106.

605 Storelli, M.M., Giacomini-Stuffler, R., Marcotrigiano, G.O., 2002. Mercury accumulation and
606 speciation in muscle tissue of different species of sharks from Mediterranean Sea, Italy.
607 *Bull. Environ. Contam. Toxicol.* 68, 201–210.

608 Suk, S.S., Smith, S.E., Ramon, D.A., 2009. Bioaccumulation of mercury in pelagic sharks from
609 the Northeast Pacific Ocean. *Cal. Coop. Ocean Fish.* 50, 172–177.

610 Teffer, A.K., 2014. Heavy metal food chain: Relating feeding ecology and mercury
611 bioaccumulation in Southern New England's top pelagic predators. Master's Thesis.
612 University of Massachusetts, Amherst, MA.

613 Trudel, M., Rasmussen, J.B., 2006. Bioenergetics and mercury dynamics in fish: a modelling
614 perspective. *Can. J. Fish Aquat. Sci.* 63, 1890–1902.

615 Torres, P., Tristão da Cunha, R., dos Santos Rodrigues, A., 2017. Mid-Atlantic elasmobranchs:
616 Suitable metal scouts? Mar. Pollut. Bull. [http://dx.doi.org/10.1016/j.marpolubul.](http://dx.doi.org/10.1016/j.marpolubul.2017.01.058)
617 2017.01.058.

618 Torres, P., Tristão da Cunha, R., dos Santos Rodrigues, A., 2016. The elasmobranch fisheries of
619 the Azores. Mar. Policy 73, 108-118.

620 Vandeperre, F., Aires-da-Silvas, A., Fontes, J., Santos, M. Serrão Santos, R., Afonso, P., 2014.
621 Movements of blue sharks (*Prionace glauca*) across their life history. PLoS ONE 9(8):
622 e103538. doi:10.1371/journal.pone. 0103538.

623 Xunta de Galicia. 2008. Ley 11/2008, del 3 de Diciembre, de pesca de Galicia. Publicado
624 en DOG núm. 243 de 16 de Diciembre de 2008 y BOE núm. 15 de 17 de Enero de 2009
625

626 **Tables**

627

628 Table 1. Number of individuals analyzed by sex, season, year and zone for blue shark and
629 shortfin mako. M: Male, F: Female.

630

631

Species	Sex		Season		Year		Zone					
	M	F	Winter	Summer	2012	2013	A	B	C	D	E	F
Blue shark	20	20	18	22	19	21	18	11	0	9	0	2
Shortfin mako	26	22	18	30	18	30	9	16	13	0	7	3

632

633

634

635

636 Table 2. Mean (SD), (*min* – *max*) total length (TL, cm), total weight (W, kg ww), total Hg
637 concentration (Hg, mg kg⁻¹ ww) and δ¹⁵N (‰) of the analyzed blue shark and shortfin mako.

638

Species	TL (cm)	W (kg ww)	Hg (mg kg ⁻¹ ww)	δ ¹⁵ N (‰)
Blue shark	160 (56) (74 – 284)	21.0 (21.2) (1.5 – 77.0)	0.52 (0.35) (0.14 – 1.71)	11.3 (0.8) (10.0 – 13.7)
Shortfin mako	156 (36) (99 – 219)	40.3 (62.7) (7.0 – 76.8)	0.74 (0.56) (0.12 – 2.57)	11.8 (0.6) (9.6 – 12.9)

639

640

641 Table 3. Mean (SD) Hg concentration (mg kg⁻¹ ww) and trophic level ($\delta^{15}\text{N}$) in the various prey
 642 of blue shark and shortfin mako. N = number of samples analyzed; unid. = unidentified

643

Prey type	N	Hg (mg kg ⁻¹ ww)	N	$\delta^{15}\text{N}$ (‰)
Cephalopods				
<i>Ancistroteuthis</i>	1	1.17	3	12.9 (0.1)
<i>lichtensteinii</i>	1	0.23	3	10.7 (0.1)
<i>Gonatus steenstrupi</i>	1	0.20	3	11.3 (0.1)
<i>Illex coindetii</i>	1	2.24	3	11.7 (0.1)
<i>Illex</i> sp.	6	1.24 (0.86)	12	12.1 (0.9)
Cephalopods unid.				
Teleost fish				
<i>Balistes capriscus</i>	1	0.06	3	10.1 (0.1)
<i>Euthynnus alletteratus</i>	2	1.08 (1.07)	6	9.7 (0.3)
<i>Scomberesox saurus</i>	8	0.08 (0.03)	12	11.2 (0.5)
<i>Scomber scombrus</i>	3	0.13 (0.08)	9	11.2 (0.2)
<i>Scomber</i> sp.	2	0.02 (0.001)	6	12.2 (0.6)
<i>Thunnus alalunga</i>	3	1.35 (0.09)	9	11.8 (0.5)
<i>Xiphias gladius</i>	1	1.43	3	12.0 (0.2)
Teleosts unid.	3	0.20 (0.21)	9	11.1 (0.1)
Chelonians				
<i>Caretta caretta</i>	1	0.59	3	9.8 (0.1)
Cetaceans				
<i>Delphinus delphis</i>	2	1.77 (0.36)	6	10.6 (0.5)
Cetaceans unid.	2	0.46 (0.05)	6	10.9 (0.1)

644

645

646

647

648 Table 4. Mean Hg concentration (mg kg⁻¹ ww) and δ¹⁵N (‰) in the two shark species and their
 649 respective diet, and biomagnification factor (BMF) calculated from sharks to their diet in all
 650 individuals combined and in the different size classes analyzed (T1-T5), N = number of
 651 individuals analyzed.

652

Species	N	Hg-shark (mg kg ⁻¹ ww)	δ ¹⁵ N-shark (‰)	Hg-diet (mg kg ⁻¹ ww)	δ ¹⁵ N-diet (‰)	BMF
Blue shark	40	0.52	11.3	0.95	11.8	0.57
T1 (<100 cm TL)	6	0.25	11.3	1.06	11.8	0.25
T2 (100-129 cm TL)	9	0.36	11.0	1.12	11.9	0.35
T3 (130-159 cm TL)	8	0.37	11.3	1.05	11.8	0.37
T4 (160-200 cm TL)	7	0.62	11.6	0.68	11.5	0.91
T5 (>200 cm TL)	10	0.86	11.3	1.03	10.9	0.80
Shortfin mako	48	0.74	11.8	0.61	11.6	1.19
T2 (100-129 cm TL)*	11	0.28	11.4	0.65	11.1	0.42
T3 (130-159 cm TL)	16	0.75	11.5	0.69	11.5	1.09
T4 (160-200 cm TL)	12	0.54	12.1	0.84	11.3	0.60
T5 (>200 cm TL)	9	1.59	12.0	0.59	10.1	2.26

653

654 * The single 99 cm TL mako was included in T2 size class

655

656

657

658

659

660

661

662

663

664

665 Table 5. Range of total Hg levels in the blue shark (*Prionace glauca*) and the shortfin mako (*Isurus*
666 *oxyrinchus*) in the Atlantic and other oceans. * = mean values; TL = total length of individuals (cm); Hg =
667 total Hg concentration in muscle (mg kg⁻¹ ww); Ref. = reference of the study cited.

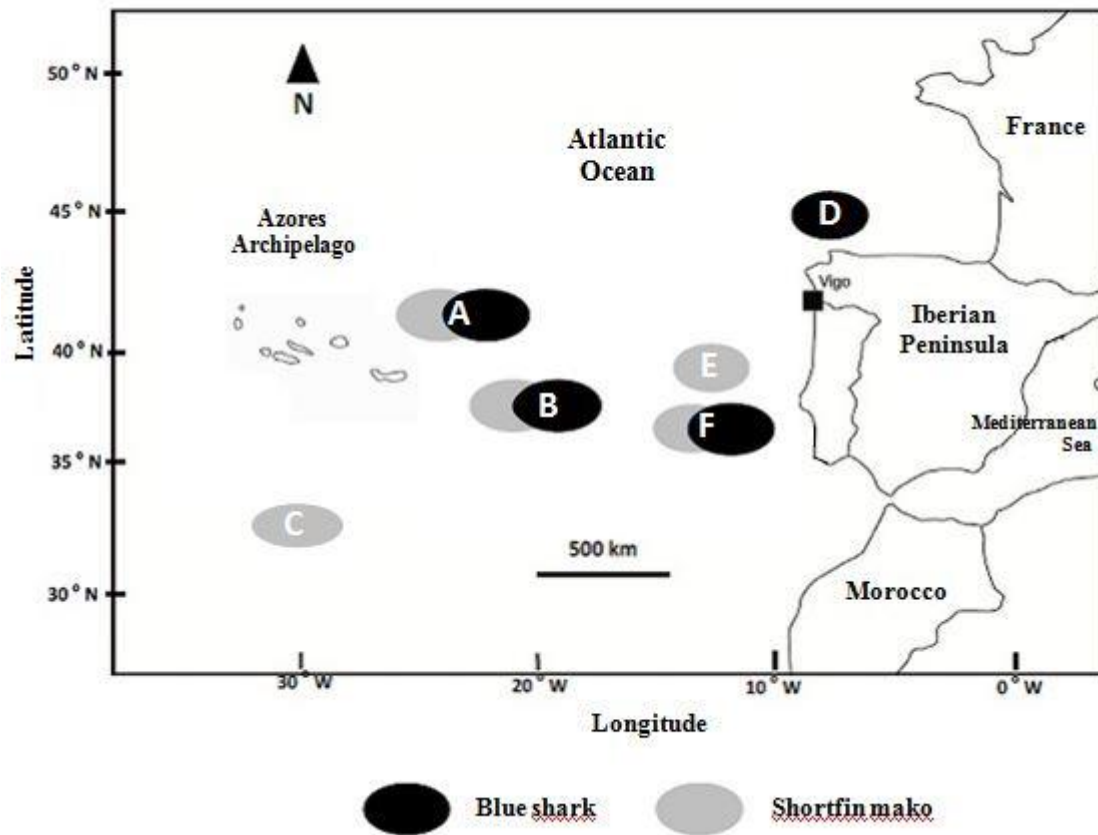
Region	Blue shark		Shortfin mako		Ref.
	TL (cm)	Hg (mg kg ⁻¹ ww)	TL (cm)	Hg (mg kg ⁻¹ ww)	
Azores & Iberian Peninsula (NE Atlantic)	79-284	0.14-1.71	100-219	0.12-2.57	(1)
Azores Archipelago (NE Atlantic)	84-239	0.16-1.84	-	-	(2, 3)
South-Eastern Azores (NE Atlantic)	98-223	0.14-0.50	153-216	0.35-1.53	(4)
Canary Islands (NE Atlantic)	216-258	0.16-1.84	-	-	(2)
New England (NW Atlantic)	-	-	199*	2.65*	(5)
North Atlantic	160-274	1.25*	106-285	3.12*	(4)
Equatorial Atlantic	172-265	0.68-2.50	-	-	(6)
South Atlantic	122-274	1.01*	94-262	2.14*	(4)
Brazil (SW Atlantic)	77-137	0.46-2.40	-	-	(7, 8)
Indian Ocean	164-269	1.24*	119-262	2.34*	(4)
South Africa (SW Indian Ocean)	-	-	161-220	2.69*	(9)
Tasmania (SW Pacific Ocean)	89-335	0.27-1.20	-	-	(10)
Baja California (NE Pacific Ocean)	-	-	127*	1.05*	(11)
California (NE Pacific Ocean)	-	-	75-330	0.15-2.90	(12)
Hawai (Pacific Ocean)	-	-	105-240	0.40-3.10	(12)
Adriatic Sea (Mediterranean)	-	0.38*	-	-	(13)

668
669 (1) Present study, (2) Branco et al. (2004), (3) Branco et al. (2007), (4) IEO (2003), (5) Teffer (2014), (6)
670 Torres et al. (2016), (7) Dias et al. (2008), (8) De Carvalho et al. (2014), (9) McKinney et al. (2016), (10)
671 Davenport (1995), (11) Maz-Courrau et al. (2012), (12), Suk et al. (2009), (13) Storelli et al. (2001).

672
673
674
675
676
677
678
679

680
681
682

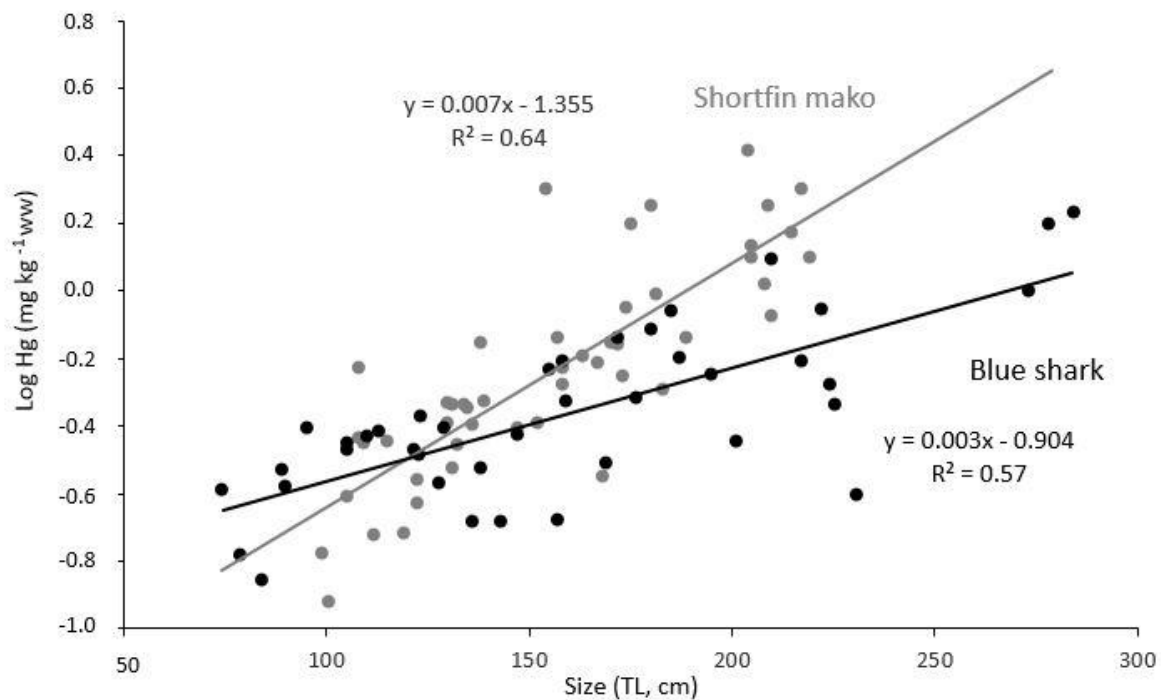
Fig. 1.



683
684
685
686
687
688
689
690
691

Fig. 1. Map of the 6 sampling areas (A to F) between the Azores Archipelago and the Iberian Peninsula (North-eastern Atlantic Ocean) and two sampled species (the blue shark in black areas and the shortfin mako in grey areas)

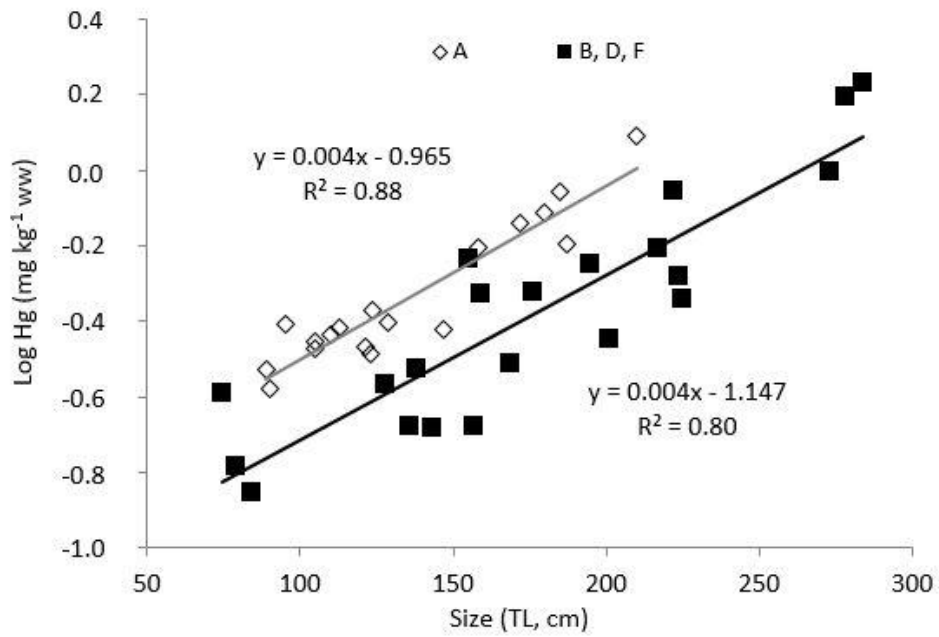
692 Fig. 2.
693



694
695 Fig. 2. Correlation between log₁₀ Hg and size in blue shark (N=40, black line and circles) and
696 shortfin mako (N=48, grey line and circles)

697
698
699
700
701
702
703
704
705

706 Fig. 3.
707



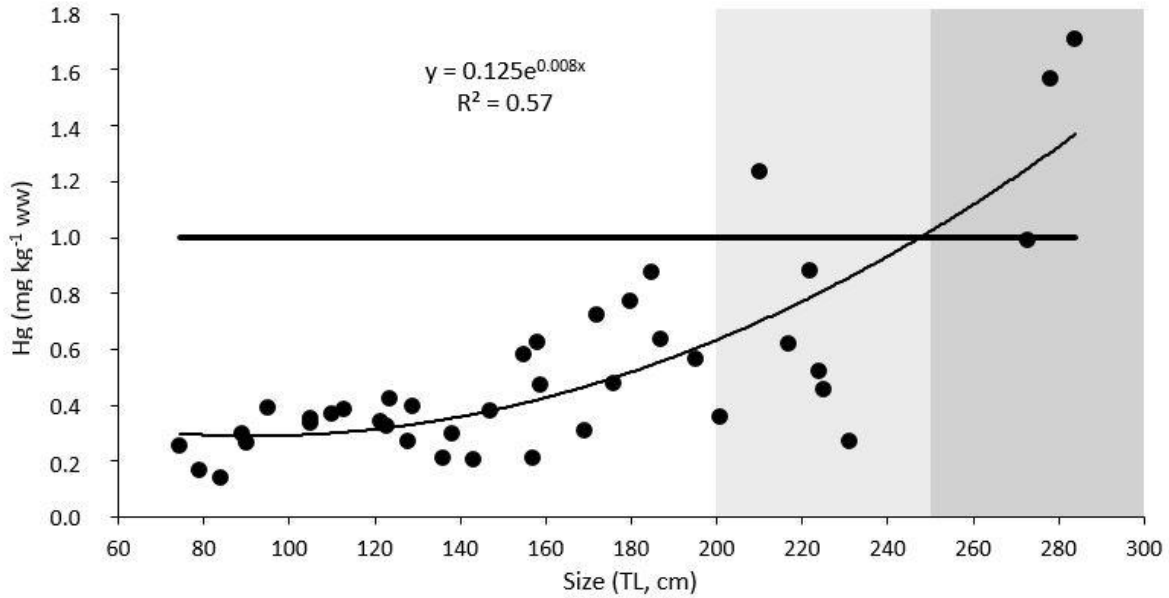
708
709

710 Fig.3. Spatial difference of correlation between log Hg and size in the blue shark in zone A
711 (northeast of the Azores, white diamonds) compared with the other sampling zones (B, D and F,
712 black squares).

713
714
715
716
717
718
719
720
721
722
723
724

725
726
727

Fig. 4.

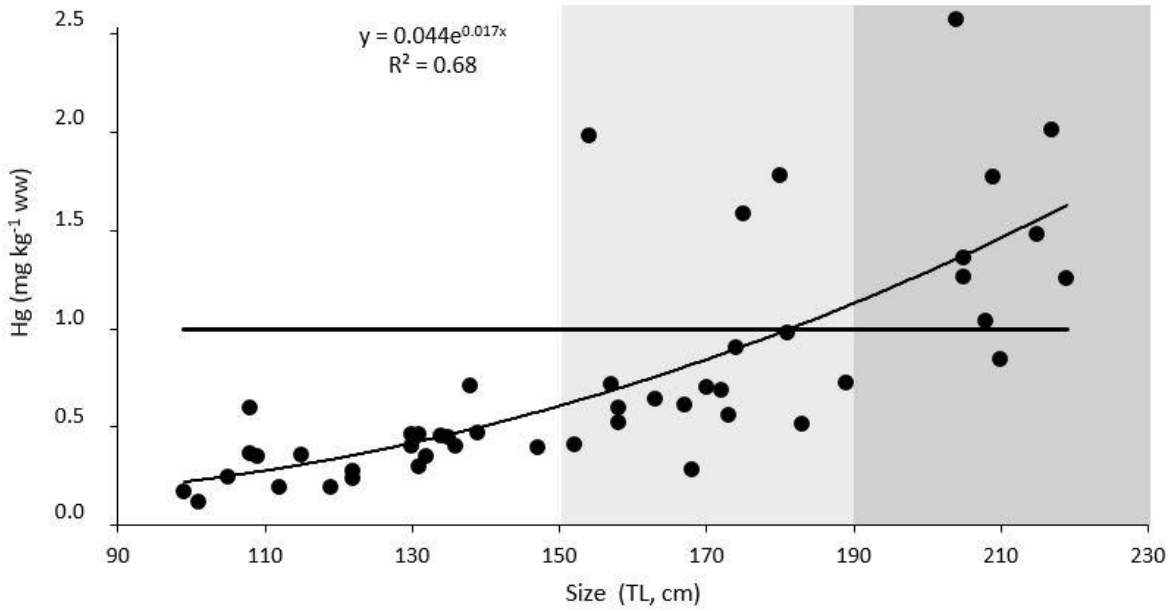


728
729
730
731
732
733
734
735
736
737
738
739
740
741
742
743
744

Fig. 4. Correlation between Hg level (mg kg⁻¹ ww) and total length (TL, cm) in the blue shark. The horizontal bold line corresponds to the European regulatory threshold (1.0 mg kg⁻¹ ww) for commercialization. The light grey area indicates the *size range of potential risk* (from which several sharks might be above the threshold) and the dark grey area starts at *the size at risk* (from which most sharks exceed the threshold).

745
746
747

Fig. 5.



748
749
750
751
752
753
754
755

Fig. 5. Correlation between Hg level (mg kg⁻¹ ww) and total length (TL, cm) in the shortfin mako. The horizontal bold line corresponds to the European regulatory threshold (1.0 mg kg⁻¹ ww) for commercialization. The light grey area indicates the *size range of potential risk* (from which several sharks might be above the threshold) and the dark grey area starts at *the size at risk* (from which most sharks exceed the threshold).