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

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Abstract :

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

Highlights

► High MeHg/THg ratio in fish muscle explained by MeHg biomagnification. ► Trophic position and feeding habitat cause Hg variation in fish muscle. ► Hg concentration was higher in muscle and liver for all fish species. ► Difference in Hg distribution could be related to differential MeHg detoxification strategies.

Keywords : Methylmercury, Contamination, Organotropism, Bioaccumulation, Teleostei

Introduction

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

species (Chouvelon et al., 2012; Cresson et al., 2015a)) and biotic factors (e.g. fish growth, trophic functioning of ecosystems (Chouvelon et al., 2018; Cossa et al., 2012; Cresson et al., 2014)). In this context, the aim of this study was to investigate Hg species (MeHg and IHg) distribution among three organs (muscle, liver, and gonad) in three commercial fish species, considering that differences in diet and feeding habitat can be potential drivers of differences.

Material and methods

Sampling area

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

Species selection and sampling collection

Three fish species were selected for their commercial interest in the Bay of Biscay, their availability regarding sampling on various habitats characteristics and different feeding habitats. Hake (*Merluccius merluccius*) is a demersal species with high commercial interest (19% of the landing value in 2017) within the Bay of Biscay (Ifremer Fisheries Information system, 2018). Canyons areas, especially the continental shelf accretion, promote concentrations of adults in the south Bay of Biscay (Alvarez et al., 2001; Sánchez and Gil, 2000; Woillez et al., 2007). Two common benthic fishes, common sole (*Solea solea*) and striped red mullet (*Mullus surmuletus*) representing 6% and 1% of the landing value respectively (Ifremer Fisheries Information system, 2018) were also selected. Sole habitat is fine and

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

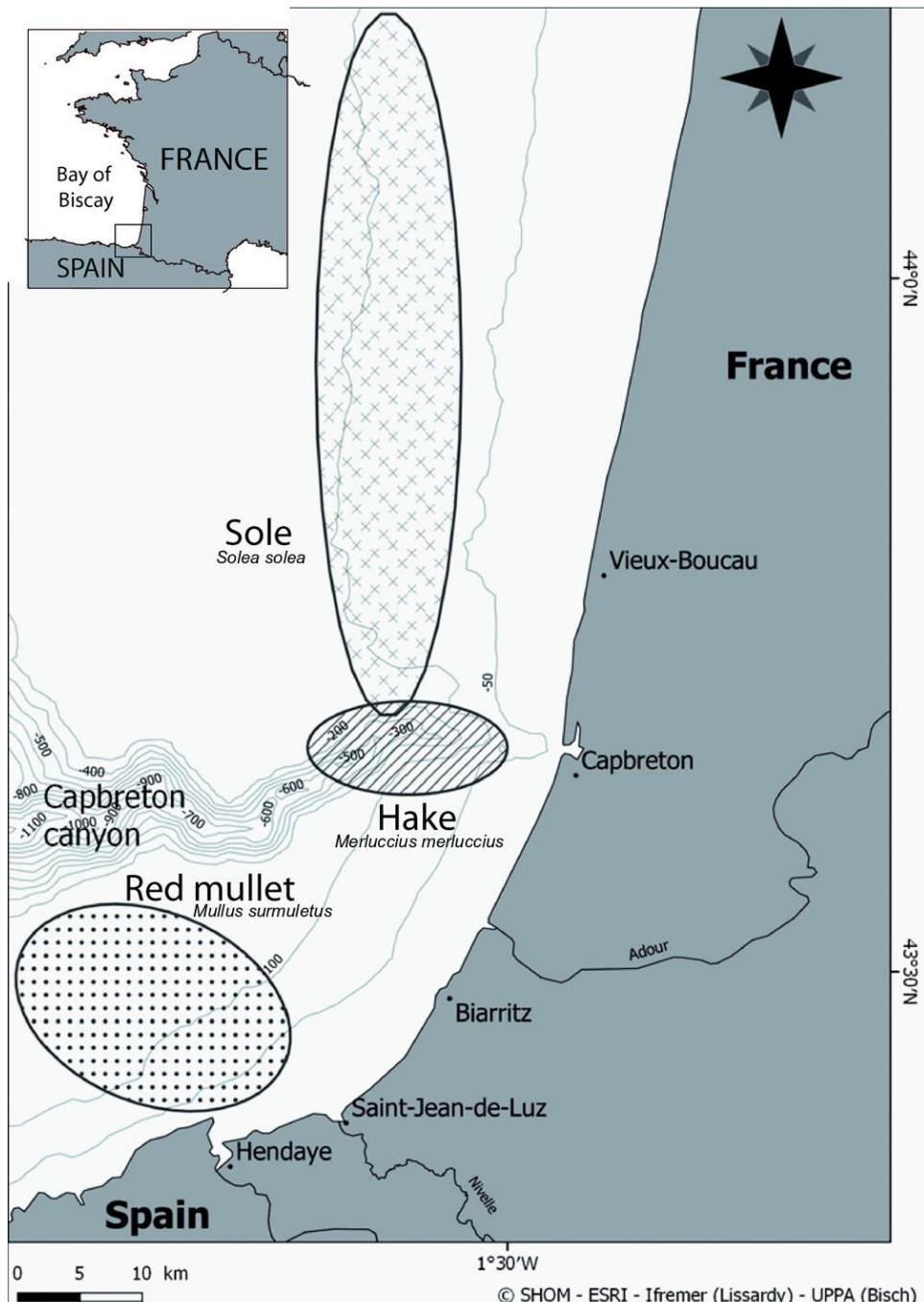


Figure 1: Map of sampling areas for the three fish species studied.

Mercury species analysis

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

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

Carbon and nitrogen stable isotope analysis

Muscle stable isotopic ratios were measured with a Thermo Scientific® Delta V Advantage mass spectrometer coupled to a Thermo Scientific® Flash EA1112 elemental analyzer following routine protocol of the Plateforme Spectrometrie Ocean (<https://www-iuem.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).

For suspended POM samples, carbon and nitrogen isotopic compositions were analyzed using an isotope ratio mass spectrometer (IRMS; Isoprime, GV Instruments®) coupled with an elemental analyzer (ThermoFisher® Scientific Flash 2000). All isotopic results are presented in the classical δ notation,

$$\delta X = \left(\frac{R_{sample}}{R_{standard}} - 1 \right) \times 10^3 \text{ where } X \text{ is } ^{13}\text{C} \text{ or } ^{15}\text{N} \text{ and } R \text{ the ratio between heavy and light isotopes.}$$

Standard is Vienna Pee Dee Belemnite for $\delta^{13}\text{C}$ and atmospheric nitrogen for $\delta^{15}\text{N}$. Carbon and nitrogen

concentration were measured with the elemental analyzer and used to calculate molar C/N ratios, classical proxy of lipid content in fish tissue for isotopic analyses. High C/N ratios are usually interpreted as high lipid content that bias isotopic measurement. All C/N ratios of fish samples were lower than 3.5 classically considered as the threshold values requiring lipid correction (Sweeting et al., 2006). Based on replicate measurements of internal laboratory standards, the analytical 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 ratios.

Statistical analysis

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

Results

Feeding ecology

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

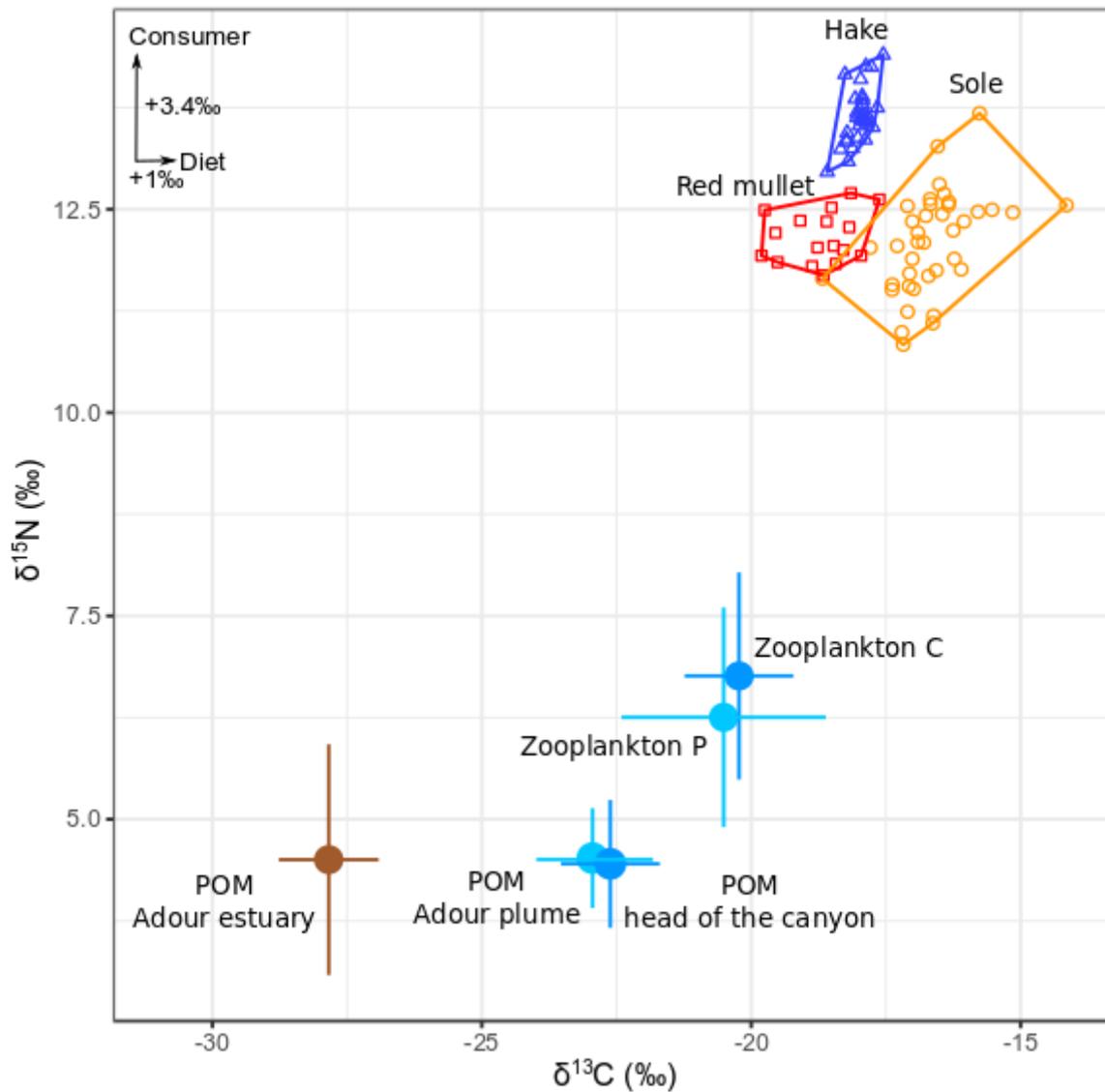


Figure 2: Relationships between carbon and nitrogen stable isotopes ratio measured in the three fish species (hake *Merluccius merluccius*, red mullet *Mullus surmuletus* and sole *Solea solea*). Signatures of particle organic matter of the Adour estuary, Adour plume, the head of the Capbreton canyon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

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 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	Go
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 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 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 5 -

Interspecies difference in mercury species concentrations in muscle

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

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

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

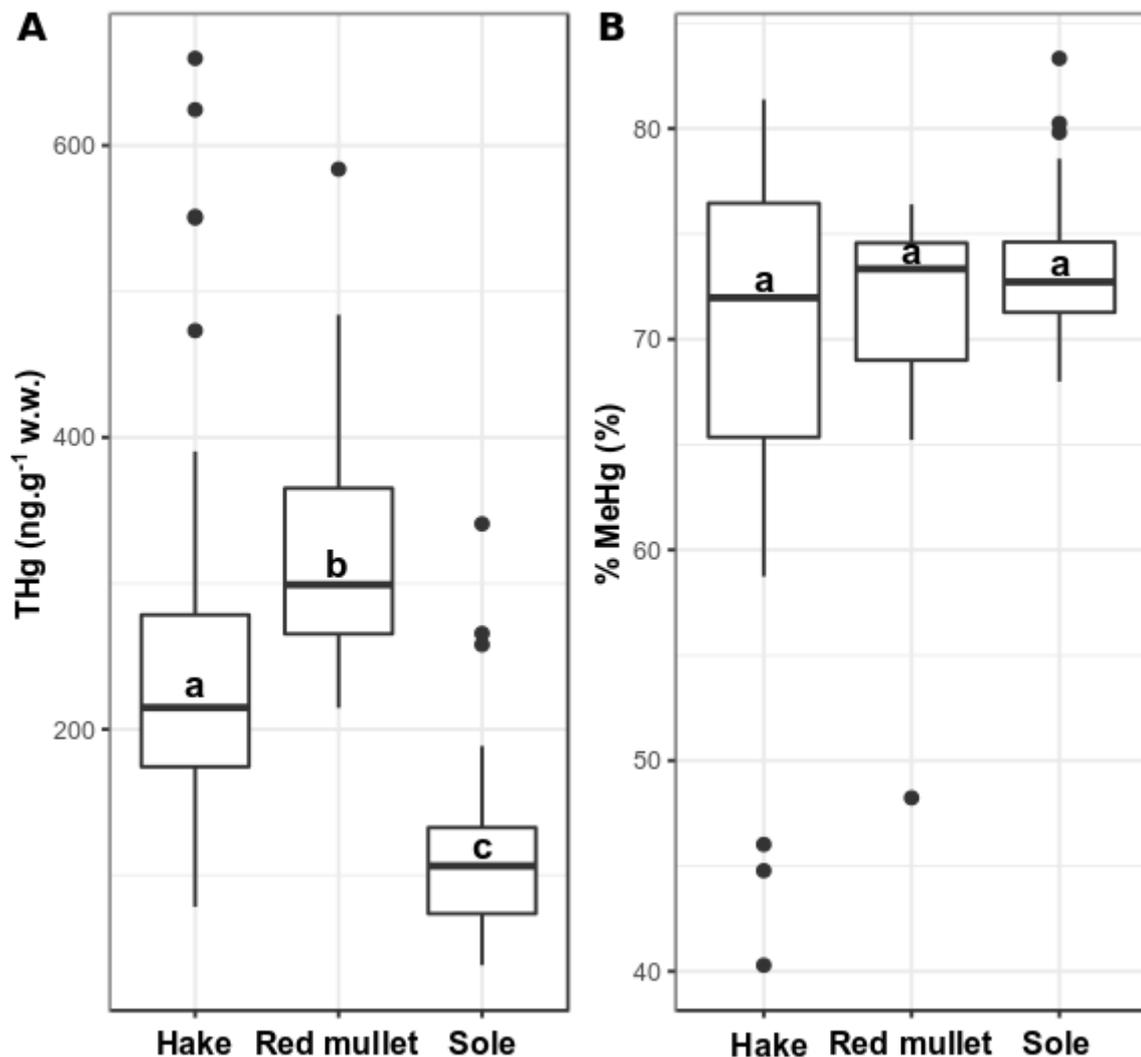


Figure 3: Total mercury concentration (A) and proportion of methylmercury (B) measured in muscle for the three species (hake *Merluccius merluccius*, red mullet *Mullus surmuletus* and sole *Solea solea*). The bottom and top of the box are the first and the third quartiles of the data distribution, the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

Distribution of mercury species between organs

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

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

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

Table 2: Differences in Hg species concentrations according to organs/tissues for the three fish species. Results were obtained with PERMANOVA illustrated 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

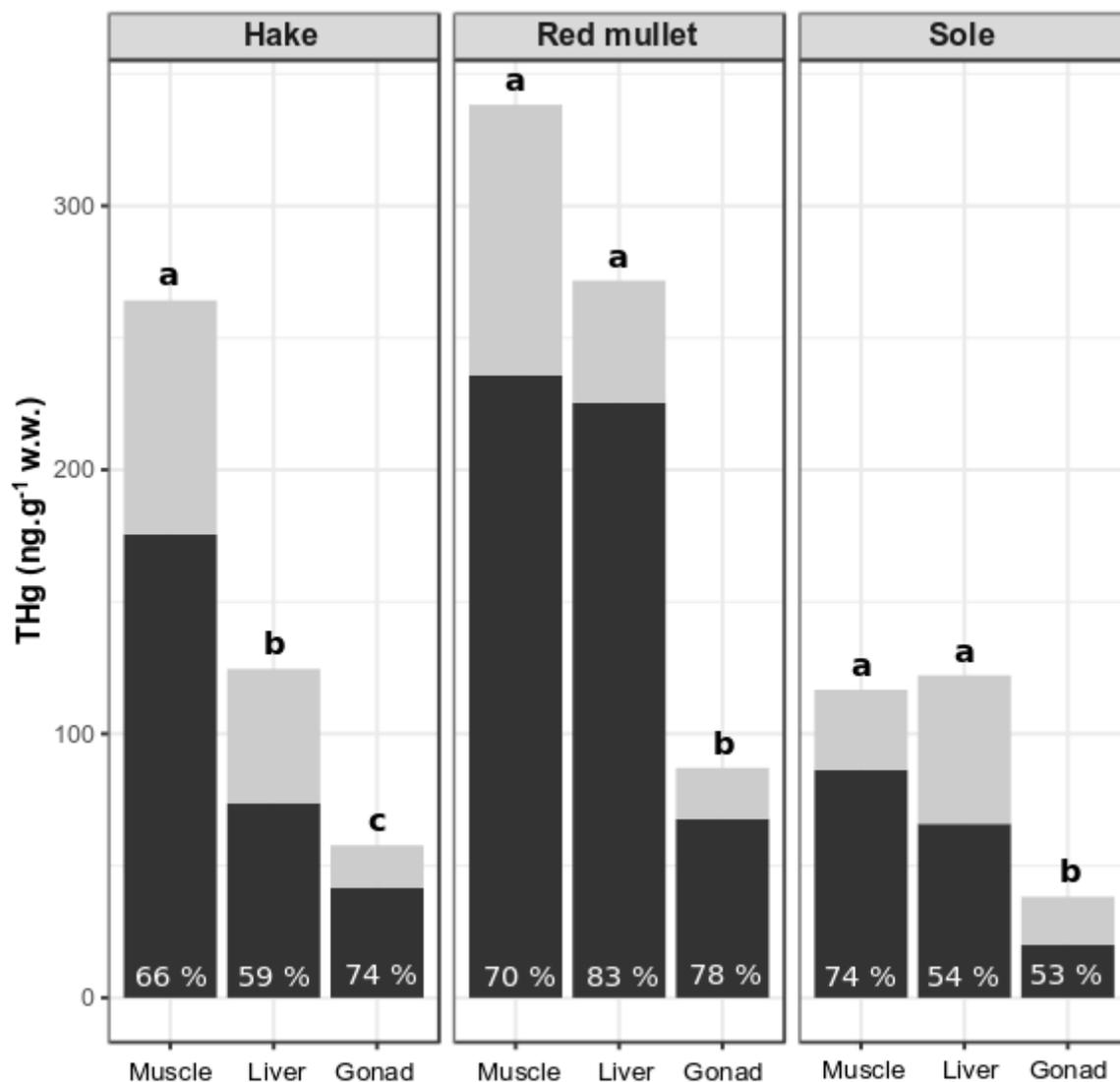


Figure 4: Distribution of Hg species according to organs for the three fish species (hake *Merluccius merluccius*, red mullet *Mullus surmuletus* and sole *Solea solea*). MeHg is displayed in black together with its percent relative to THg, and IHg is in grey. Standard deviation are not added for graphical purposes. Significant differences between organs are highlighted with different letters.

Discussion

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

The present study confirmed that MeHg represent the majority of Hg in fish, consistently with most literature records (Chouvelon et al., 2018; Pethybridge et 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 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 can exhibit cannibalism and consume young conspecifics (Mahe et al., 2007; Pinnegar, 2014; Velasco and Olaso, 2000). Red mullet and sole

are benthic omnivorous fish consuming small crustaceans, worms and mollusks including small bivalves (Caill-Milly et al., 2017; Le Loc'h et al., 2008). The pattern observed for nitrogen isotopic ratio, with higher values for hake, is thus consistent the expected relative trophic position of the three fish species. Therefore, 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 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 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 variability of Hg concentration. Fish position in the water column was also an important factor, since pelagic species presented significantly lower THg concentration in muscle than the benthic species (Chouvelon et al., 2012). This pattern is exemplified here, as red mullet had highest THg concentrations than 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 Capbreton canyon, a presumed sink for organic matter, sediment and contaminants especially Hg (Azaroff et al., 2019; Gaudin et al., 2006). This pattern was also observed in the Mediterranean, where fish living in canyons were more contaminated than counterparts living away (Cresson et al., 2016; Koenig et al., 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 canyon that was less contaminated in Hg than offshore (Azaroff et al., 2019). Isotopic values measured for hake are consistent with values previously 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 this species. It may thus be assumed that they poorly use benthic organic matter from the canyons, but fed above on the pelagic pathway.

Despite having similar $\delta^{15}\text{N}$ values, red mullet presented significantly higher THg concentrations than sole. This difference may result from differences in 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 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 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

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 pathway. On the rocky shore substrate where mullets were caught, pelagic production can enter directly in the food web after falling, since bacterial processes 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 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 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 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 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 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.* 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 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, difference in trophic level may originate difference in Hg concentrations. Overall, this study confirms the importance of food sources within the feeding habitat/ecosystem as a major driver of Hg bioaccumulation in marine species muscles (Chouvelon et al., 2018; Cresson et al., 2015b).

Differential Hg species distribution within organs

As expected, Hg concentrations were higher in muscle and liver. This organotropism of Hg is consistent with other studies about Hg distributions in organs (Kasper et al., 2009; Polak-Juszczak, 2018). Biochemical characteristics of fish tissues, fish metabolism and Hg chemical behavior are the main drivers of 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 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 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, hake presented a significant

difference of Hg concentrations between muscle and liver. These results suggested different capacities or physiological ability of 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 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 MeHg to selenium or proteins (*e.g.* glutathione and metallothionein), resulting a less harmful chemical form (Penglase et al., 2014). The lowest %MeHg in sole liver could trace an increase importance of this pathway. Several studies demonstrated that flatfish such as the four-spot megrim *Lepidorhombus boscii* developed increased detoxication abilities, as a response to their chronic exposure to contaminant from diet and sediment (García et al., 2000; Martínez-Gómez et al., 2006). Therefore, differences in Hg distribution between tissues can be attributed to species related to physiological processes especially the detoxification strategies involved in Hg elimination and/or biotransformation (Le Croizier et al., 2018).

The gonads are composed mainly by lipoglycophosphoprotein, because teleost oocytes are mainly composed by vitelline proteins. Therefore, the proportion of 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 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 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 transfer a portion of their accumulated contaminants to their offspring during reproduction (Govoni et al., 2017; Lyons and Lowe, 2013; Stefansson et al., 2014). Spawning was thus interpreted as a depuration process in Hg for females, in some freshwater fish species (Donald and Sardella, 2010; Hammerschmidt et al., 1999; Johnston et al., 2001).

Conclusions

In summary, this study has investigated and compared Hg species concentrations in the muscle of three commercial fish species. All individuals except four hakes had THg concentrations in muscle above the food safety thresholds determined by the European Commission regulation. We conclude that the distribution of Hg species (organic and inorganic) is fish species specific. Therefore, differences in MeHg bioaccumulation according to fish species are consequences of their diet, their metabolism

(especially MeHg demethylation of processes) and their feeding habitat. Results of the present study confirms the importance of considering the environment in which they reside to investigate Hg bioaccumulation in marine fish. A larger sample size involving larger individuals could be considered, to confirm the results presented above and thus strengthen the European monitoring program.

Acknowledgements

The research leading to these results has received funding from the ERDF (European Regional Development Fund) and AEAG (Agence de l'Eau Adour-Garonne) under the MICROPOLIT project. Authors are grateful to Maud Ezan who helped in samples preparation and to Muriel Lissardy who drawn the sampling map. Thanks are due to teams of the Plateforme de Spectrometrie Océan (Rudolph Corvaisier and Oanez Lebeau) and of the Plateforme ISOTOPIE of EPOC (Nicolas Savoye and Karine Charlier) who performed the isotopic analyses.

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Electronic supplementary Material

Table S1: Length and sex effect on Hg species concentrations according to organs for the three commercial fish species. Results were obtained with a linear regression and illustrated by the F statistic together with the corresponding p-value (bold for significant value). A log transformation on concentration data proved necessary for the linear regressions in order to achieve the assumption of normality and homogeneity of variance that were checked on residual data.

		Muscle		Liver		Gonad	
		MHg	IHg	MHg	IHg	MHg	IHg
Hake <i>Merluccius merluccius</i>	Length	F = 5.35, p.value = 0.028	F = 0.30, p.value > 0.005	F = 1.77, p.value > 0.005	F = 0.88, p.value > 0.005	F = 0.55, p.value > 0.005	F = 0.61, p.value > 0.005
	Sex	F = 0.83, p.value > 0.005	F = 0.30, p.value > 0.005	F = 0.69, p.value > 0.005	F = 3.34, p.value > 0.005	F = 0.18, p.value > 0.005	F = 1.68, p.value > 0.005
Red mullet <i>Mullus surmuletus</i>	Length	F = 0.23, p.value > 0.005	F = 0.39, p.value > 0.005	F = 0.03, p.value > 0.005	F = 0.20, p.value > 0.005	F = 0.12, p.value > 0.005	F = 0.72, p.value > 0.005
	Sex	F = 2.22, p.value > 0.005	F = 0.56, p.value > 0.005	F = 0.34, p.value > 0.005	F = 0.69, p.value > 0.005	F = 0.01, p.value > 0.005	F = 5.90, p.value > 0.005
Sole <i>Solea solea</i>	Length	F = 2.70, p.value > 0.005	F = 5.08, p.value > 0.005	F = 0.01, p.value > 0.005	F = 0.02, p.value > 0.005	F = 0.01, p.value > 0.005	F = 0.52, p.value > 0.005
	Sex	F = 8.92, p.value = 0.005	F = 5.93, p.value = 0.020	F = 2.56, p.value > 0.005	F = 6.69, p.value = 0.014	F = 3.01, p.value > 0.005	F = 17.55, p.value = 0.001

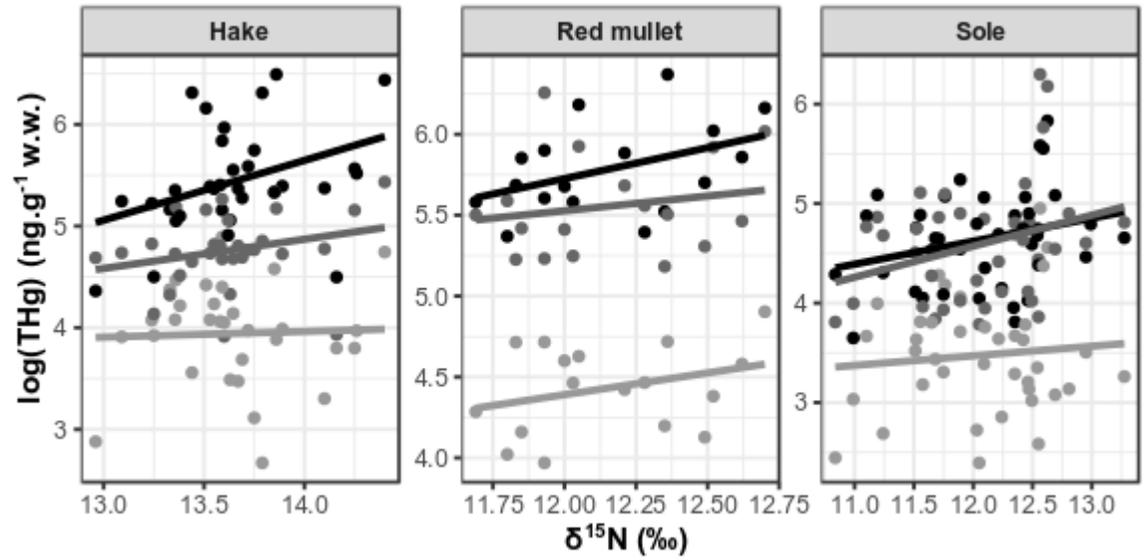
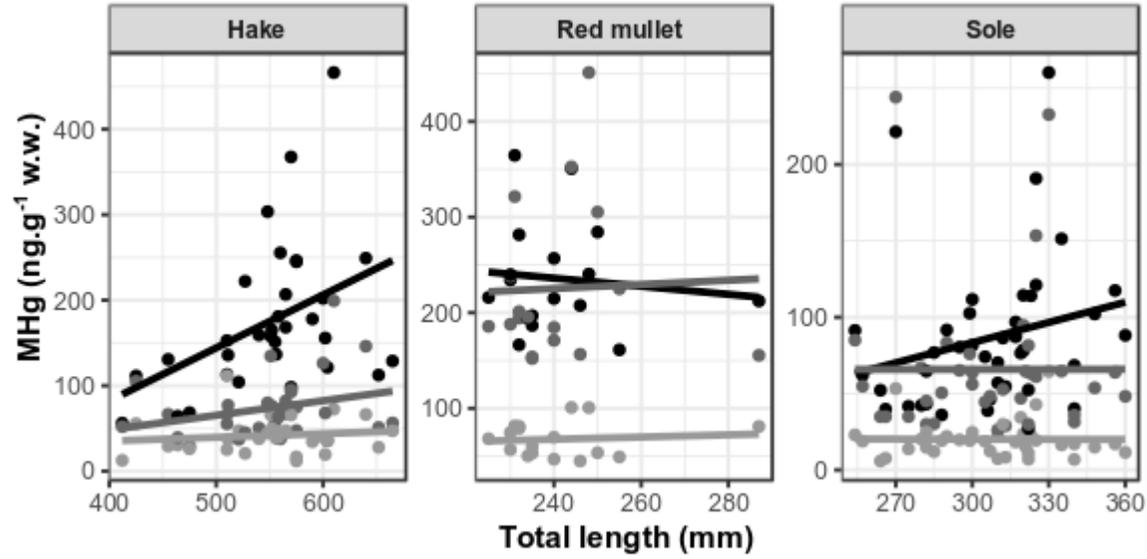


Figure S1: Relationship between MHg and total length (upper row) and relationship between log THg and trophic level (lower row). Colors stand for the organs or tissues considered (muscle in black, liver in medium grey and gonad in light grey) for the three fish species.

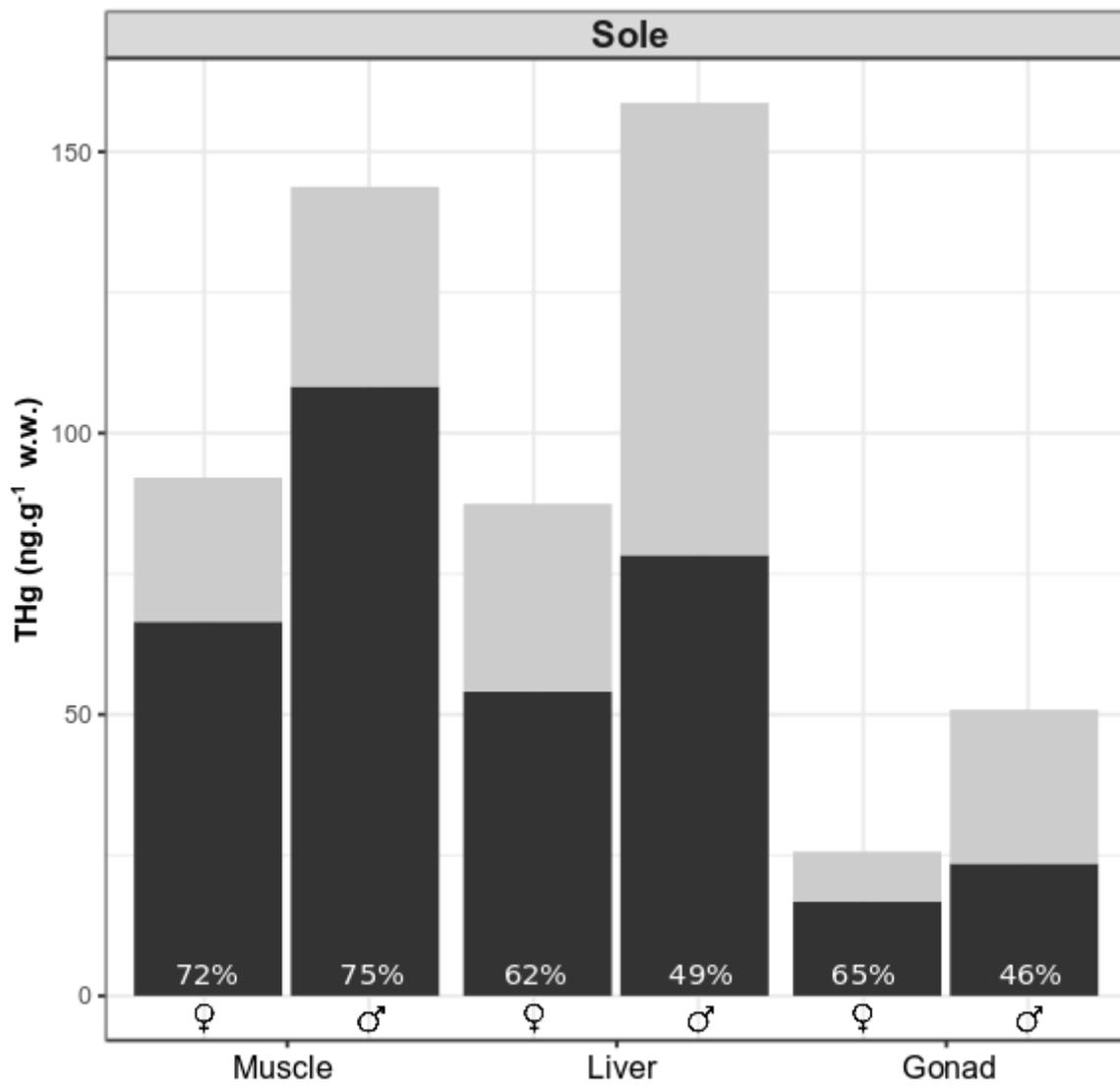


Figure S2: Hg distribution according to organs/tissues and sex for sole *Solea solea* (MeHg in black together with %MeHg, and IHg in grey). Standard deviation are not added for graphical purposes.