Environmental Pollution 233 (2018) 844–854

Contents lists available at ScienceDirect

Environmental Pollution

journal homepage: www.elsevier.com/locate/envpol

Oligotrophy as a major driver of mercury bioaccumulation in medium-to high-trophic level consumers: A marine ecosystem-comparative study☆

Tiphaine Chouvelon [a, *], Pierre Cresson [b, c], Marc Bouchoucha [b], Christophe Brach-Papa [a, b], Paco Bustamante [d], Sylvette Crochet [a], Françoise Marco-Miralles [b], Bastien Thomas [a], Joël Knoery [a]

[a] IFREMER, Unité Biogéochimie et Écotoxicologie, Laboratoire de Biogéochimie des Contaminants Métalliques, Rue de l'Ile d'Yeu, BP 21105, 44311 Nantes Cedex 03, France
[b] IFREMER, Unité Littoral, Laboratoire Environnement Ressources Provence Azur Corse, Zone portuaire de Brégaillon, CS 20330, 83507 La Seyne sur Mer Cedex, France
[c] IFREMER, Unité Halieutique de Manche-Mer du Nord, Laboratoire Ressources Halieutiques de Boulogne, 150 quai Gambetta, 62200 Boulogne sur Mer, France
[d] Littoral Environnement et Sociétés (LIENSs), UMR 7266 CNRS-Université de La Rochelle, 2 rue Olympe de Gouges, 17042 La Rochelle Cedex 01, France

ARTICLE INFO

Article history:
Received 2 August 2017
Received in revised form
2 October 2017
Accepted 4 November 2017

Keywords:
Biomagnifying metal
Bioaccumulation
Trophic status
Ecosystem productivity
Temperate systems

ABSTRACT

Mercury (Hg) is a global contaminant of environmental concern. Numerous factors influencing its bioaccumulation in marine organisms have already been described at both individual and species levels (e.g., size or age, habitat, trophic level). However, few studies have compared the trophic characteristics of ecosystems to explain underlying mechanisms of differences in Hg bioaccumulation and biomagnification among food webs and systems. The present study aimed at investigating the potential primary role of the trophic status of systems on Hg bioaccumulation and biomagnification in temperate marine food webs, as shown by their medium-to high-trophic level consumers. It used data from samples collected at the shelf-edge (i.e. offshore organisms) in two contrasted ecosystems: the Bay of Biscay in the North-East Atlantic Ocean and the Gulf of Lion in the North-West Mediterranean Sea. Seven species including crustaceans, sharks and teleost fish, previously analysed for their total mercury (T-Hg) concentrations and their stable carbon and nitrogen isotope compositions, were considered for a meta-analysis. In addition, methylated mercury forms (or methyl-mercury, Me-Hg) were analysed. Mediterranean organisms presented systematically lower sizes than Atlantic ones, and lower $\delta^{13}C$ and $\delta^{15}N$ values, the latter values especially highlighting the more oligotrophic character of Mediterranean waters. Mediterranean individuals also showed significantly higher T-Hg and Me-Hg concentrations. Conversely, Me-Hg/T-Hg ratios were higher than 85% for all species, and quite similar between systems. Finally, the biomagnification power of Hg was different between systems when considering T-Hg, but not when considering Me-Hg, and was not different between the Hg forms within a given system. Overall, the different parameters showed the crucial role of the low primary productivity and its effects rippling through the compared ecosystems in the higher Hg bioaccumulation seen in organisms from oligotrophic Mediterranean waters.

1. Introduction

Mercury (Hg) is released into the environment from both natural and anthropogenic sources (e.g., volcanism, fuel and waste combustion). It reaches marine waters through atmospheric depositions and riverine inputs, the former being the main source of Hg for the surface ocean (Fitzgerald et al., 2007). Also, Hg is volatile

☆ This paper has been recommended for acceptance by Dr. Yong Sik Ok.

* Corresponding author.
E-mail address: tiphaine.chouvelon@ifremer.fr (T. Chouvelon).

https://doi.org/10.1016/j.envpol.2017.11.015
0269-7491/

and as such it can be transported through the atmosphere over long distances and deposited in areas away from its point sources. In the ocean, trophic transfer represents the main pathway for the intake and bioaccumulation of Hg by marine organisms. Hg exhibits a specific behaviour compared to other trace metals, as it biomagnifies through food webs in its organic methyl-Hg (Me-Hg) forms (Chen et al., 2008). Finally, Hg is of particular environmental concern because of its toxicity on the nervous, reproductive, immunological and hormonal systems, sometimes leading to harmful effects at the population level (Boening, 2000; Tan et al., 2009; Goutte et al., 2014).

The methylation of Hg by microorganisms into Me-Hg is the fundamental process giving this contaminant its bioaccumulation and biomagnification properties. In the marine environment, Hg methylation occurs both in coastal and shelf sediments, and in subthermocline low-oxygen oceanic waters, at depths where organic matter is intensively remineralized (Mason et al., 1995; Fitzgerald et al., 2007; Blum et al., 2013; Cossa et al., 2017). In its methylated forms, Hg passes through biological membranes easily and is incorporated into cellular cytoplasm (e.g., in phytoplankton), from which it is assimilated very efficiently by higher trophic levels (Le Faucheur et al., 2014). Once incorporated in organisms, Hg strongly binds with protein sulfhydryl groups (−SH). Due this affinity, the elimination or the excretion of the bioaccumulated Hg is very slow over time, sometimes equal to zero, like in the muscle tissues (Wang and Wong, 2003; Maulvault et al., 2016). As a consequence, Hg has been widely documented to bioaccumulate with size or age in marine organisms (e.g., Monteiro and Lopes, 1990; Cossa et al., 2012; Chouvelon et al., 2014a).

Several factors linked to Hg marine biogeochemical cycling and its chemical properties are known to influence Hg bioaccumulation in marine fauna. The concentrations of dissolved Me-Hg in ambient waters or in the different habitats of species is a first factor. As an example, mesopelagic species were shown to present higher Hg concentrations than epipelagic ones (Monteiro et al., 1996; Choy et al., 2009; Chouvelon et al., 2012), due to the probably higher exposition of mesopelagic organisms to Me-Hg in these deep-pelagic layers of organic matter remineralisation (e.g., Cossa et al., 2009; Heimbürger et al., 2010). Time of exposure to Hg is a second factor that directly influences its bioaccumulation. It is generally inferred by the size or the age of the organisms considered, and usually higher Hg concentrations are measured in older organisms (due to low elimination rates; see above). The trophic status and/or trophic functioning of ecosystems is a third factor that may be put forward to explain different bioaccumulation rates (e.g., Chen and Folt, 2005). This “trophic factor” itself includes several aspects, some being intimately linked to the first two described above. The first aspect of this trophic factor directly concerns system productivity, through the “bio-dilution effect” (Pickhardt et al., 2002). The bio-dilution effect suggests lower Hg bioaccumulation at all trophic levels in mesotrophic areas compared to oligotrophic areas (Harmelin-Vivien et al., 2009; Cresson et al., 2014a). It is due to the higher number and higher surface area/volume ratio (i.e. size) of cells at the base of mesotrophic systems. This configuration is less favourable to an efficient uptake of Hg by cells, whose the lower Hg burden (in comparison with cells in oligotrophic area) is then transferred to consumers. Moreover in oligotrophic areas, where cells are thus less abundant and potentially contain higher Hg burden, primary consumers probably consume virtually all of them. A second aspect of the trophic factor is directly related to the occurrence of specific planktonic communities at the base of food webs. Indeed, in oligotrophic environments, the higher proportions of slowly sinking pico- and nanophytoplanctonic cells that are readily degraded by bacteria within the water column promote Me-Hg formation in the heterotrophic active layers of organic matter remineralisation (e.g., Cossa et al., 2009; Heimbürger et al., 2010). This aspect is thus intimately derived from the first factor concerning the level of bioavailable Hg in ambient waters. A third aspect of the trophic factor concerns the “trophic chain length”, also corresponding to the number of “nodes” between the considered organisms and the primary producers. The trophic chains are sometimes considered to be longer in less productive environments (i.e. due to the higher recycling of organic matter and bacterial activity; e.g., Biddanda et al., 2001). In the case of biomagnifying contaminants such as Hg, it may therefore lead to higher Hg concentrations measured in apex predators of oligotrophic systems. Finally, we may hypothesize that the trophic status and associated primary productivity of marine systems influence the growth rate of organisms as well (i.e. lower grow rates in less productive or oligotrophic environments may be expected), hence also influencing the rate of Hg bioaccumulation (e.g., Simoneau et al., 2005; Trudel and Rasmussen, 2006; Ward et al., 2010).

Following these three fundamental factors (i.e. environmental concentrations of Me-Hg in ambient waters; age or growth rate of the organisms considered in the different systems; trophic status of systems), which can be related to each other, the bioaccumulation and the biomagnification of Hg were proved to vary sometimes greatly between marine ecosystems (e.g., Harmelin-Vivien et al., 2009; Cossa et al., 2012), or even between environments of a same marine ecosystem (e.g., between epipelagic and mesopelagic environments; Choy et al., 2009; Chouvelon et al., 2012; Cresson et al., 2014a). For instance, the Mediterranean Sea was found to be a peculiar marine ecosystem, characterized by several factors favourable to Hg contamination yielding to the so-called “Mediterranean Hg anomaly”, whereby organisms' Hg concentrations reported are often higher in this sea than in others while seawater concentrations are comparable (Cossa and Coquery, 2005; Harmelin-Vivien et al., 2009; Cossa et al., 2012). However, relatively few ecosystem-comparative or meta-analysis studies exist for assessing the relative influence of the different factors described above (e.g, Lavoie et al., 2013), and the probable primary role of oligotrophy on Hg bioaccumulation; especially considering offshore species (i.e. expected to be more affected by oligotrophic conditions than neritic species in any system), and/or considering several species at a time (and not only a single species). Moreover, lake ecosystems are more documented (e.g., Chen and Folt, 2005; Kidd et al., 2012; Lavoie et al., 2013) than the marine environment (but see Harmelin-Vivien et al., 2009; Cossa et al., 2012). Finally, the reasons for among-systems differences in Hg bioaccumulation and biomagnification have been recently described as being yet not well understood, especially those relative to ecosystems' characteristics (Kidd et al., 2012; Lavoie et al., 2013).

In this context, the general objective of this study was to compare and evaluate the influence of the trophic status fo systems and of biological processes (i.e. over geochemical ones; e.g., Cossa and Coquery, 2005) on Hg bioaccumulation and biomagnification in offshore species and food webs from the Bay of Biscay (BoB) in the north-eastern (NE) Atlantic, and from the Gulf of Lions (GoL) in the north-western (NW) Mediterranean. The species included benthopelagic crustaceans and fish (both teleost and cartilaginous) that are characteristic of the shelf-edge in both ecosystems. They were selected to avoid potential bias linked to direct water-discharge inputs of Hg and/or coastal processes on Hg cycling and bioaccumulation. Besides, the BoB in the NE Atlantic and the GoL in the NW Mediterranean represent good candidates for such comparative study, firstly due their expected difference in terms of trophic status, even offshore (one being open onto the Ocean, the other being a semi-enclosed Sea). Indeed, the NE Atlantic shelves (such as the Bay of Biscay) and the Mediterranean Sea are considered distinct biogeochemical provinces for a long time (e.g.,

Longhurst, 1998, 2007; Reygondeau et al., 2013). Moreover, the more oligotrophic character of the Mediterranean system compared to the Atlantic one has been previously documented, especially from the composition of the organic matter (e.g., presence of diazotrophic organisms) sustaining food webs (Kerhervé et al., 2001; Liénart et al., 2017). Nonetheless, a number of species can be found in both ecosystems, allowing direct comparisons. Finally, both areas are major areas for fisheries (for which Hg bioaccumulation in commercial species may be of concern), and they constitute two out the three French façades considered and monitored by the European Marine Strategy Framework Directive.

To fulfil its general objective, this ecosystem-comparative study used comparable and analytically coherent data on total Hg (T-Hg) concentrations and carbon (C) and nitrogen (N) stable isotope ratios (as markers of food sources, trophic positions, and ecosystems' properties in terms of trophic status) analysed on the selected species, as described separately by Chouvelon et al. (2012) for the BoB and by Cresson et al. (2014b) for the GoL. In addition, for the present study, analyses of Me-Hg were performed on the samples, to investigate whether the percentage of Me-Hg measured in the selected species may differ between systems. Indeed, while Me-Hg represents the bioaccumulated form of Hg, it is rarely measured and is often considered to be nearly equal to 100% of total Hg (T-Hg), although this can vary between taxa and species (Bustamante et al., 2006; Kehrig et al., 2010; Cossa et al., 2012; Briant et al., 2017). Finally, the present additional analysis of Me-Hg allowed comparing the biomagnification rates of T-Hg and Me-Hg within each system, and between the two contrasted systems for each Hg form. Overall, we hypothesized that the trophic status and especially the oligotrophic character of Mediterranean waters should strongly influence the bioaccumulation and/or biomagnification of Hg observed in medium-to high-trophic level consumers, due to the lower productivity and consequently the lower growth rate and lower "bio-dilution effect" most probably generated at all trophic levels in oligotrophic systems.

2. Material and methods

2.1. Sampling and sample preparation

The offshore (shelf-edge) benthopelagic species considered in this study included the lesser-spotted dogfish *Scyliorhinus canicula* and the blackmouth catshark *Galeus melastomus* as cartilaginous fish; the blackbelly rosefish *Helicolenus dactylopterus*, the four-spot megrim *Lepidorhombus boscii*, the greater forkbeard *Phycis blennoides* and the blue whiting *Micromesistius poutassou* as teleost fish; and finally the Norway lobster *Nephrops norvegica* as crustacean (Table 1). Organisms were collected during bottom-trawling groundfish surveys conducted by the French Institute for the Exploitation of the Sea (IFREMER), in 2008 for the BoB and in 2012 for the GoL (Fig. 1; Chouvelon et al., 2012; Cresson et al., 2014b).

After collection (N = 134 fishes in total considered in the present meta-analysis and comparative study), each individual was measured (total length for fish, cephalothorax length for crustaceans), at least to nearest centimetre for fish or millimetre for crustaceans. A piece of white muscle (without skin nor carapace) was taken for both Hg analyses and C and N stable isotope analysis (SIA), performed individually. All muscle samples were finally frozen at −20 °C, freeze-dried and ground into a fine powder until further chemical analyses (Chouvelon et al., 2012; Cresson et al., 2014b).

The determination of the age of organisms could not be performed due to non-uniform sampling of otoliths for age lecture, although in any case, this age determination would have been only potentially possible for teleost fish (that is, for four of the seven species considered here), if the otolith sampling had been done uniformly. Thus, in the present study, only the individual sizes were used as an indicator of the time of exposure to contaminant (i.e. as a proxy of organisms' age within a species), and/or related to potential differences in the growth rate of all organisms between systems.

2.2. Stable isotope analyses

Muscle sub-samples for SIA (N = 134) were prepared as described by Chouvelon et al. (2012) and Cresson et al. (2014b). 0.40 ± 0.05 mg of powder were finally weighed in tin cups. Analyses were performed with a Thermo Scientific Delta V Advantage mass spectrometer coupled to a Thermo Scientific Flash EA1112 elemental analyser. The results are presented in the usual δ notation relative to the deviation from international standards (Pee Dee Belemnite for $\delta^{13}C$ values, and atmospheric nitrogen for $\delta^{15}N$ values), in parts per thousand (‰). Based on replicate measurements of internal laboratory standards (acetanilide), the experimental precision was <0.2‰ for both $\delta^{13}C$ and $\delta^{15}N$. Moreover, a previous study using the same instruments as in the present study indicated no statistically significant difference for both $\delta^{13}C$ and $\delta^{15}N$ on duplicate measurements of a same sample (Chouvelon et al., 2014b), indicating highly reliable and precise values for the unique analysis of well-homogenized material such as the muscle samples used here.

2.3. Total and methyl-Hg analyses

As in the case of SIA, muscle sub-samples for T-Hg determination (N = 134) were prepared and analysed as described by Chouvelon et al. (2012) and Cresson et al. (2014b). T-Hg analyses were carried out on aliquots of powder (10–50 mg) with Advanced Mercury Analysers (ALTEC AMA-254, Altec Ltd). Through this mean, the determination of Hg is done in three steps: (1) combustion of the dedicated sub-sample (dried powder) and volatilization of Hg; (2) amalgamation of elemental Hg on a gold trap; and (3) spectrophotometric atomic absorption measurement of the Hg swept into the flow cell following heating (800 °C) of the gold trap. All T-Hg analyses were run according to thorough quality control programmes including the analysis of certified reference materials (CRMs): TORT-2 (lobster hepatopancreas, National Research Council Canada/NRCC), IAEA-142 (mussel homogenate, International Atomic Energy Agency/IAEA), DORM-2 and/or DORM-4 (fish protein, NRCC). CRM results were in good agreement with the certified values in all cases, with recovery rates varying between 95% and 102% (Chouvelon et al., 2012; Cresson et al., 2014b). The limit of quantification was ≤0.02 μg g^{-1} dry mass (dm).

Me-Hg determination was performed on a subsample of individuals (n = 75) according to the method described by Azemard and Vassileva (2015), which uses liquid-liquid extractions of Me-Hg in samples before analysis by automated Hg analysers such as AMA-254. Briefly, aliquots of powder (20–800 mg, depending on T-Hg concentrations) were acidified with 5 mL of HCl (25%, v/v, prepared with HCL 30% Suprapur) to solubilize Hg. A volume of toluene (10 mL) was added and both phases were homogenized. After centrifugation, a fraction (5 mL) of the upper organic phase (i.e. toluene containing extracted Me-Hg) was transferred to a second tube containing the same volume of a 0.002 M sodium thiosulfate solution. This second tube was vigorously shaken and centrifuged. Finally, an aliquot of a known volume (100 or 200 μL) of the lower phase, containing the back-extracted Me-Hg, was directly analysed with AMA 254. As for T-Hg determination, Me-Hg analyses were run according to a thorough quality control programme including the analysis of the CRM IAEA-436 (tuna fish flesh homogenate). The

Table 1
Characteristics of fish and crustaceans considered in the NW Mediterranean (Gulf of Lion – GoL) and in the NE Atlantic (Bay of Biscay – BoB): sizes (in cm), stable isotope ratios ($\delta^{13}C$ and $\delta^{15}N$ values in ‰) and muscle total Hg (T-Hg) concentrations (in $\mu g\ g^{-1}$ dm) for the individuals analysed for the three parameters (N = 134); sizes (in cm), methyl-Hg (Me-Hg) concentrations (in $\mu g\ g^{-1}$ dm) and MeHg/T-Hg ratio (in %) for the subsample of individuals analysed for Me-Hg concentrations (n = 75). The water content (WC) of the muscle tissue (in %), either derived from dedicated individual measures (for five of the seven Atlantic species) or from the general literature (US EPA, 2011; in brackets) is also indicated, for further conversion of Hg concentrations in wet mass if needed (i.e. for comparison with seafood Hg standards). (nd) = not determined.

Ecosystem	N	Size [a] Mean ± SD (min, max)	$\delta^{13}C$ Mean ± SD (min, max)	$\delta^{15}N$ Mean ± SD (min, max)	T-Hg Mean ± SD (min, max)	WC, measured Mean ± SD (min, max)	WC, US EPA 2011 Mean ("Raw" tissue) [b]	n	Size [a] (subsampling) Mean ± SD (min, max)	Me-Hg Mean ± SD (min, max)	Me-Hg/T-Hg ratio Mean ± SD (min, max)
NW Mediterranean (GoL)	**72**							**37**			
Cartilaginous fish											
Scyliorhinus canicula (Sc)	9	48 ± 5 (41, 57)	−18.0 ± 0.2 (−18.4, −17.8)	8.9 ± 0.2 (8.7, 9.3)	10.85 ± 7.62 (4.63, 27.01)	(nd)	(73.6)	6	50 ± 5 (41, 57)	11.61 ± 8.45 (4.95, 27.07)	99 ± 5 (93, 107)
Galeus melastomus (Gm)	15	46 ± 6 (33, 56)	−18.0 ± 0.3 (−18.4, −17.2)	8.9 ± 0.6 (7.8, 10.2)	7.81 ± 4.02 (3.37, 16.72)	(nd)	(73.6)	6	48 ± 3 (43, 51)	8.59 ± 3.94 (3.44, 13.25)	101 ± 7 (91, 112)
Teleost fish											
Helicolenus dactylopterus (Hd)	13	26 ± 3 (20, 31)	−18.3 ± 0.5 (−19.0, −17.5)	9.2 ± 0.8 (7.9, 10.2)	7.83 ± 2.97 (4.24, 12.92)	(nd)	(79.3)	6	27 ± 3 (22, 31)	7.20 ± 3.26 (3.49, 11.46)	93 ± 7 (82, 103)
Lepidorhombus boscii (Lb)	5	26 ± 5 (20, 33)	−18.6 ± 0.2 (−18.9, −18.4)	8.2 ± 0.3 (7.9, 8.6)	4.96 ± 2.66 (2.33, 9.30)	(nd)	(79.1)	4	28 ± 4 (24, 33)	5.13 ± 3.36 (1.89, 9.59)	94 ± 12 (81, 105)
Phycis blennoides (Pb)	15	27 ± 5 (20, 38)	−18.8 ± 0.5 (−19.8, −17.9)	9.4 ± 0.7 (7.1, 10.1)	2.63 ± 1.03 (1.14, 4.21)	(nd)	(81.2)	6	29 ± 4 (23, 34)	2.77 ± 0.57 (2.25, 3.84)	93 ± 2 (90, 96)
Micromesistius poutassou (Mp)	12	26 ± 3 (22, 31)	−19.0 ± 0.6 (−20.5, −18.3)	8.6 ± 0.4 (8.0, 9.4)	1.53 ± 0.45 (0.90, 2.41)	(nd)	(80.3)	6	26 ± 3 (23, 31)	1.36 ± 0.56 (0.74, 2.27)	86 ± 5 (81, 94)
Crustaceans											
Nephrops norvegicus (Nn)	3	4.7 ± 0.4 (4.3, 5.0)	−18.8 ± 0.4 (−19.1, −18.3)	7.0 ± 0.3 (6.7, 7.2)	3.11 ± 0.50 (2.69, 3.66)	(nd)	(76.8)	3	4.7 ± 0.4 (4.3, 5.0)	2.86 ± 0.51 (2.38, 3.40)	92 ± 3 (89, 94)
NE Atlantic (BoB)	**62**							**38**			
Cartilaginous fish											
Scyliorhinus canicula (Sc)	10	58 ± 3 (53, 63)	−16.7 ± 0.2 (−16.9, −16.3)	13.1 ± 0.3 (12.6, 13.5)	2.12 ± 1.19 (0.93, 4.63)	77.0 ± 0.6 (76.0, 78.2)	(73.6)	6	57 ± 2 (54, 60)	1.98 ± 0.74 (1.18, 3.03)	94 ± 4 (88, 98)
Galeus melastomus (Gm)	12	61 ± 7 (50, 72)	−17.2 ± 0.2 (−17.7, −16.8)	12.1 ± 0.6 (11.2, 13.2)	2.19 ± 1.38 (1.04, 5.12)	78.3 ± 0.4 (77.7, 78.9)	(73.6)	6	63 ± 8 (52, 72)	2.51 ± 1.56 (0.95, 4.53)	94 ± 6 (89, 105)
Teleost fish											
Helicolenus dactylopterus (Hd)	5	37 ± 2 (34, 40)	−17.3 ± 0.1 (−17.5, −17.2)	13.2 ± 0.3 (12.7, 13.6)	4.77 ± 0.84 (3.89, 6.13)	79.6 ± 0.6 (78.7, 80.2)	(79.3)	5	37 ± 2 (34, 40)	4.87 ± 0.81 (4.09, 6.19)	102 ± 4 (99, 107)
Lepidorhombus boscii (Lb)	5	30 ± 3 (26, 34)	−16.8 ± 0.1 (−17.0, −16.6)	11.5 ± 0.3 (11.2, 11.9)	2.32 ± 1.80 (0.82, 4.29)	(nd)	(79.1)	5	30 ± 3 (26, 34)	1.94 ± 1.40 (0.79, 3.56)	88 ± 7 (79, 97)
Phycis blennoides (Pb)	5	51 ± 7 (44, 58)	−17.0 ± 0.3 (−17.4, −16.7)	13.5 ± 0.1 (13.3, 13.6)	0.96 ± 0.72 (0.36, 1.80)	(nd)	(81.2)	5	51 ± 7 (44, 58)	0.97 ± 0.75 (0.35, 1.83)	99 ± 4 (93, 104)
Micromesistius poutassou (Mp)	20	25 ± 5 (20, 33)	−18.1 ± 0.4 (−19.1, −17.1)	11.5 ± 0.6 (10.1, 13.0)	0.25 ± 0.22 (0.08, 0.77)	78.4 ± 0.9 (76.7, 79.8)	(80.3)	6	26 ± 5 (21, 32)	0.30 ± 0.28 (0.07, 0.66)	95 ± 6 (84, 103)
Crustaceans											
Nephrops norvegicus (Nn)	5	6.4 ± 0.7 (5.7, 7.4)	−15.9 ± 0.2 (−16.2, −15.6)	11.3 ± 0.2 (11.1, 11.5)	0.62 ± 0.07 (0.55, 0.69)	78.3 ± 1.3 (76.9, 80.0)	(76.8)	5	6.4 ± 0.7 (5.7, 7.4)	0.59 ± 0.06 (0.51, 0.64)	94 ± 2 (92, 96)

[a] Total length for fish, cephalothorax length for crustaceans.
[b] Selected correspondence (closest) species in US EPA (2011): *S. canicula* and *G. melastomus* = "Sharks, mixed species"; *H. dactylopterus* = "Rockfish"; *L. boscii* = "Flatfish, Flounder, and Sole"; *P. blennoides* = "Cod, Atlantic"; *M. poutassou* = "Whiting, mixed species"; *N. norvegicus* = "Lobster, northern".

average recovery rates for the CRM varied from 81 to 90% between the series of analyses (i.e. between the days of analyses), against 92% on average expected by Azemard and Vassileva (2015) for this CRM. However, within a same series or day of analyses, the variation in the recovery rates of CRMs (2–3 CRMs analysed at each series/day of Me-Hg analyses) was very low (i.e. average standard deviation of 2%). Therefore, the repeatability of the CRM results was ensured within a same series, and Me-Hg results for the samples could be corrected from the average recovery rate of the CRMs analysed during the same series/day of analyses. Finally, several samples were analysed twice (in two different series/day of analyses). The mean difference observed between the two measurements, for the calculated percentage of Me-Hg relative to T-Hg (i.e. ratio of Me-Hg concentration/T-Hg concentration, in %), was of 4 ± 2%.

2.4. Data treatment

All data submitted to statistical tests (and/or model residuals) were checked for normality, and for homogeneity of variances and lack of violation of independence when appropriate (Zuur et al., 2007).

For each species considered, differences between systems in individual sizes, $\delta^{13}C$ and $\delta^{15}N$ values, T-Hg concentrations and percentages of Me-Hg were thus tested by the parametric Student t-test or the non-parametric Mann-Whitney-Wilcoxon test, depending on whether the data satisfied the conditions for parametric statistics or not.

The correlation between T-Hg and Me-Hg concentrations (for the subsample of individuals analysed for both parameters) was tested through the non-parametric Spearman correlation coefficient test.

The relationship between T-Hg or Me-Hg concentrations and $\delta^{15}N$ values (i.e. as proxy of the individual trophic positions within a given ecosystem) was investigated through Generalized Linear Models (GLMs). Indeed, data showed a marked departure from normality, preventing the application of classical multiple linear regressions. However, thorough data exploration suggested the linearity of trends to be modelled, making the application of Generalized Additive Models – than can capture and model complex non-linear relationships – unnecessary. GLMs were thus fitted to log-transformed Hg concentrations with an identity link

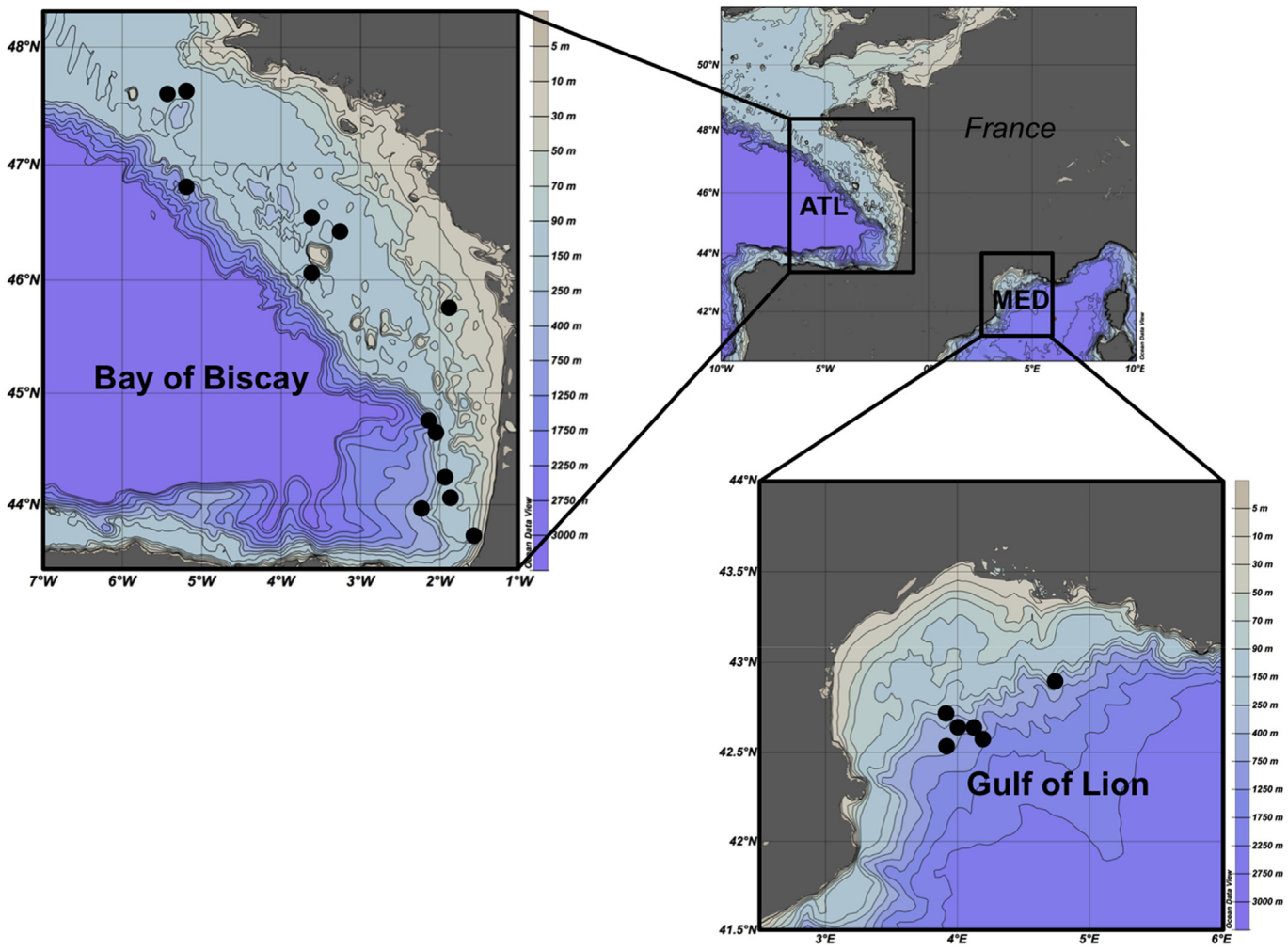

Fig. 1. Maps of the sampling areas and of trawling stations in the NW Mediterranean (MED – Gulf of Lion) and in the NE Atlantic (ATL – Bay of Biscay).

function, as it is generally the case when dealing with contaminant data such as trace metals (e.g., Pierce et al., 2008; Mèndez-Fernandez et al., 2013; Chouvelon et al., 2014a, 2017). This allowed estimating the effect of $\delta^{15}N$ values in explaining Hg concentration variability. Moreover, such relationships allowed the biomagnification power (BP) of T-Hg and Me-Hg to be determined and to be compared between ecosystems (Chen et al., 2008; Borgå et al., 2012; Lavoie et al., 2013). $\delta^{15}N$ values were treated as the single continuous explanatory variable in the models, while the factor "Ecosystem" was treated as a categorical explanatory variable and added as a potential interaction term. The general form of the original models performed was:

Log_{10} [T-Hg] or Log_{10} [Me-Hg] ~ $\delta^{15}N$ + Ecosystem [+ interaction term $\delta^{15}N$:Ecosystem].

Results (fitted values) of the models were plotted on observed (log-transformed) data. Final models' parameters (estimates, p-values, etc.) and details on their interpretation are given in Table 2. For each model, we retained the variables that improved the relative goodness of fit in the GLMs (i.e. most parsimonious models) based on the Akaike Information Criterion (AIC). When the AIC was not significantly different between the last two nested models, the simplest model was preferred. Finally, a model validation was systematically applied (Zuur et al., 2007), and the percentage of total deviance explained (DE) was calculated as follows: Explained deviance = ((Null model deviance - final model residual deviance)/Null model deviance)*100, with the null model that only contained the intercept terms (Mèndez-Fernandez et al., 2013; Chouvelon et al., 2017).

The same type of models (GLMs) was finally applied to test potential significant difference in the slope of the relationships (i.e. in the BP) between T-Hg and Me-Hg (i.e. between the different forms of Hg) within each system. $\delta^{15}N$ values were treated as the single continuous explanatory variable in the models, while the factor "Hg form" was treated as a categorical explanatory variable and added as a potential interaction term. The general form of the original models performed was:

Log_{10} [Hg] in the BoB or in the GoL ~ $\delta^{15}N$ + Hg form [+ interaction term $\delta^{15}N$:Hg form].

The level of significance for statistical analyses was always set at $\alpha = 0.05$.

3. Results

In both systems, species-dependent patterns in Hg concentrations (either T-Hg or Me-Hg) were similar, with the two shark species *S. canicula* and *G. melastomus* and the teleost fish

Table 2
Results of the final GLM models explaining (log-transformed) T-Hg or Me-Hg concentrations' variability in the muscle of the seven selected species. Akaike Information Criterion (AIC) values and the total deviance explained (DE) by each model are indicated (see section 2.4). Estimates and significance (p-values) for each term included are also given. *p < 0.05; **p < 0.01; ***p < 0.001.

	Variables (equation terms) [a]	Estimates	p-value
T-Hg (N = 134)	**Log10 [T-Hg] = δ15N + Ecosystem + δ15N:Ecosystem**		
	AIC = 138.8		
	Total DE = 53.8%		
	Intercept	(α) −4.4378	<0.001***
	$\delta^{15}N$	(β_1) 0.3573	<0.001***
	MED-GoL (relative to ATL-BoB)	4.0925	<0.001***
	$\delta^{15}N$:MED-GoL (relative to $\delta^{15}N$:ATL-BoB)	−0.2484	0.0034**
Me-Hg (n = 75)	**Log10 [Me-Hg] = δ15N + Ecosystem**		
	AIC = 78.0		
	Total DE = 44.7%		
	Intercept	(α) −2.5701	<0.001***
	$\delta^{15}N$	(β_1) 0.2136	<0.001***
	MED-GoL (relative to ATL-BoB)	0.9858	<0.001***

[a] When the interaction term is not significant (i.e. model assuming that the form of the relationship between ecosystems is the same, for Me-Hg), the underlying model's specifications are:
$Log_{10}\,[Me\text{-}Hg]_i = \alpha + \beta_1 *\delta^{15}N_i + Ecosystem_i + \varepsilon_i$ (1)
When the interaction is significant (i.e. for T-Hg, form of the relationship different between ecosystems), the model's specifications are:
$Log_{10}\,[T\text{-}Hg]_i = \alpha + \beta_1 *\delta^{15}N_i + Ecosystem_i + \delta^{15}N_i{:}Ecosystem_i *\delta^{15}N_i + \varepsilon_i$ (2)
In equations (Eqs. (1) and (2)), the terms correspond to:
- Log_{10} [T-Hg or Me-Hg]$_i$ = the log-transformed concentration in T-Hg or Me-Hg for sample i;
- α = intercept for the relationship;
- β_1 = the estimate for the continuous explanatory variable $\delta^{15}N$;
- $\delta^{15}N_i$ = the $\delta^{15}N$ value for sample i;
- Ecosystem$_i$ = correction to apply for the ecosystem(s) of concern if necessary, here for MED-GoL (see below);
- ε = residuals (i.e. information not explained by the model).
When the interaction is not significant in the model (i.e. for Me-Hg), predicted values (fitted model) derived from model outputs can thus be deduced as follow:
- Log_{10} [Me-Hg]$_i$ for ATL-BoB (i.e. first modality of the factor Ecosystem) = $\alpha + \beta_1 *\delta^{15}N_i$
- Log_{10} [Me-Hg]$_i$ for MED-GoL (i.e. second modality of the factor Ecosystem) = $\alpha + \beta_1 *\delta^{15}N_i$ + estimate for MED-GoL
When the interaction is significant, it is in the form (i.e. for T-Hg):
- Log_{10} [T-Hg]$_i$ for ATL-BoB = $\alpha + \beta_1 *\delta^{15}N_i$
- Log_{10} [T-Hg]$_i$ for MED-GoL = $\alpha + \beta_1 *\delta^{15}N_i$ + estimate for MED-GoL + estimate for the interaction term $\delta^{15}N$:MED-GoL $*\delta^{15}N_i$
In fact, in the model outputs of predicted values, the estimates for the first modality of the factor Ecosystem (i.e. ATL-BoB) are equal to 0 because the model uses this first modality as baseline (Zuur et al., 2007). Estimates for the modality MED-GoL thus correspond to the correction applying to this modality, relative to the modality ATL-BoB.

H. dactylopterus presenting the highest Hg concentrations, the two teleost fish *P. blennoides* and *L. boscii* presenting intermediate concentrations, and the teleost fish *M. poutassou* and the crustacean *N. norvegica* presenting the lowest Hg concentrations (Table 1).

Mediterranean individuals had significantly lower sizes than those from the NE Atlantic (BoB) for five out of the seven species considered (Table 1, Fig. 2a), along with significantly higher T-Hg concentrations for all species except *L. boscii* (Table 1, Fig. 2b). However, although non significant for *L. boscii*, higher average values were found in Mediterranean individuals of this species as well (Table 1). Concomitantly, for all species considered, significantly lower $\delta^{13}C$ and $\delta^{15}N$ values were measured in Mediterranean organisms relative to Atlantic ones (Table 1, Fig. 2c).

Me-Hg concentrations were strongly correlated with T-Hg concentrations (Fig. 3), and the average percentage of Me-Hg (relative to T-Hg) was over 85% for all the considered species and in both systems (Table 1, Fig. 2d). This percentage of Me-Hg was significantly higher in Atlantic individuals relative to Mediterranean ones for only three out of the seven species considered (i.e. for *H. dactylopterus*, *P. blennoides* and *M. poutassou*), although slightly, and the difference was not significant for the four other species (Fig. 2d). Moreover, the standard deviations around average Me-Hg/T-Hg ratios were quite high (Table 1), and the recovery of values between individuals from the different systems was quite important (Fig. 2d).

The effect of $\delta^{15}N$ values for explaining T-Hg or Me-Hg concentrations' variability was significant in all cases (i.e. significant relationships between parameters), as well as the factor "Ecosystem" (i.e. significant difference between Atlantic and Mediterranean). The intercept was systematically higher for Mediterranean organisms than for Atlantic ones in all relationships (Table 2, Fig. 4). For Me-Hg, contrary to T-Hg, the interaction term was not significant. As such, the model predicted similar slope/similar BP for Me-Hg in both ecosystems, at least at the scale of the species and food webs considered in the present study. Finally, when considering each system separately, the difference of slope/of BP between T-Hg and Me-Hg was not significant (i.e. no effect of the factor "Hg form" on the relationships with $\delta^{15}N$ values within each system), probably due to the high correlation between T-Hg and Me-Hg concentrations (Fig. 3).

4. Discussion

4.1. General trends and differences between systems

Results clearly showed a significantly higher Hg bioaccumulation by Mediterranean organisms, along with significantly lower C and N stable isotope ratios measured for all species, and lower individual sizes for most of them. Conversely, the percentage of MeHg was similar between both systems for most of the species examined, although significantly slightly higher in Atlantic organisms than in Mediterranean ones for some species. When considering Me-Hg, the BP was not significantly different between systems, but was significantly higher in the Atlantic food web than in the Mediterranean one when considering T-Hg. Besides, within each system, T-Hg and Me-Hg BPs appeared to not differ significantly.

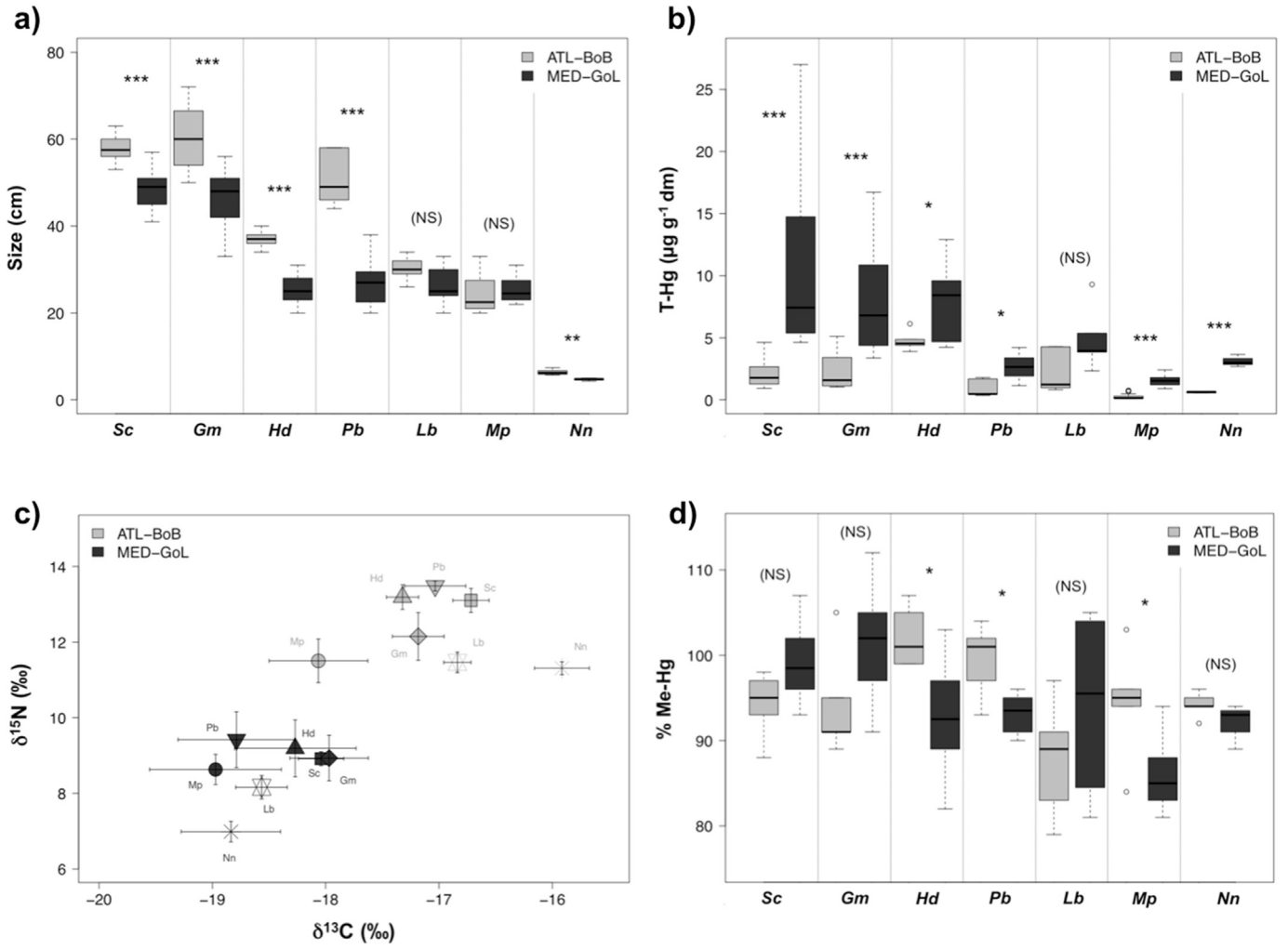

Fig. 2. a) Boxplots of sizes (total length for fish, cephalothorax length for crustaceans) for the seven species analysed in the NW Mediterranean – Gulf of Lion (MED – GoL) and in the NE Atlantic – Bay of Biscay (ATL – BoB) (N = 134); b) Boxplots of muscle total Hg (T-Hg) concentrations (N = 134); c) Biplot of $\delta^{13}C$ and $\delta^{15}N$ values (N = 134); d) Boxplots of percentages of Me-Hg (i.e. ratios Me-Hg/T-Hg, in %) in the muscle of the subsample of individuals analysed for Me-Hg concentrations (n = 75).
For boxplots (a, b, d), the box length represents the interquartile, the bar length represents the range, and the horizontal lines in bold are median values. For each species, the significant difference between systems is indicated (tested by Student t-test or Mann-Whitney-Wilcoxon test, depending on data satisfying conditions for parametric statistics or not). $^{*}p < 0.05$; $^{**}p < 0.01$; $^{***}p < 0.001$; NS = non significant. For the biplot (c), values are mean ± standard deviation per species. Abbreviations for species are specified in Table 1.

Firstly, in terms of T-Hg concentrations, the species-dependent pattern observed was similar between systems. Moreover, it was consistent with the general diet and ecology documented for the considered species (www.sealifebase.org and associated references) and with the well-documented biomagnifying property of Hg in food webs (Boening, 2000). Indeed, the two shark species *S. canicula* and *G. melastomus* and the teleost fish *H. dactylopterus* (i.e. species presenting the highest Hg concentrations) are considered high-trophic level consumers mainly feeding on a wide variety of prey, including both benthic and pelagic fish, cephalopods and crustaceans. Compared with the others, these three species are also morphologically able to capture relatively large prey (i.e. of potentially high trophic level as well). Alternatively, *P. blennoides* is documented to feed mostly on crustacean and fish prey, and *L. boscii* on small bottom-living crustaceans, fish and squids. The diet of the individuals of *M. poutassou* corresponding to the sizes sampled here is mainly composed of small crustaceans such as zooplankton. Finally, the crustacean *N. norvegica* is considered mainly scavenger, feeding on detritus, benthic crustaceans and worms (www.sealifebase.org and associated references).

Secondly, the different parameters considered in this ecosystem-comparative study (i.e. sizes of the organisms compared, $\delta^{13}C$ and $\delta^{15}N$ values, T-Hg and Me-Hg concentrations) showed the crucial role of the trophic status at the base of systems – especially oligotrophy – in the trophic transfer and in the bioaccumulation of Hg by high-trophic level consumers. Similarly to our study, higher concentrations in Mediterranean organisms than in their counterparts in the Atlantic were recently described for the neritic food web of the European hake *Merluccicus merluccius* between the GoL and the BoB (Cossa et al., 2012), being coherent with the "Mediterranean Hg anomaly" (i.e. higher Hg bioaccumulation observed in organisms from this system relative to other systems, despite comparable concentrations in seawaters from both areas; Aston and Fowler, 1985; Cossa and Coquery, 2005). However, to the best of our knowledge, such comparison of systems as done in the present study was relatively rarely performed, especially on several medium-to high-trophic level species at a time, and on offshore species, which are potentially submitted to oligotrophic conditions – at least in part – in both systems (compared to neritic species).

The C and N stable isotope compositions (especially $\delta^{15}N$ values)

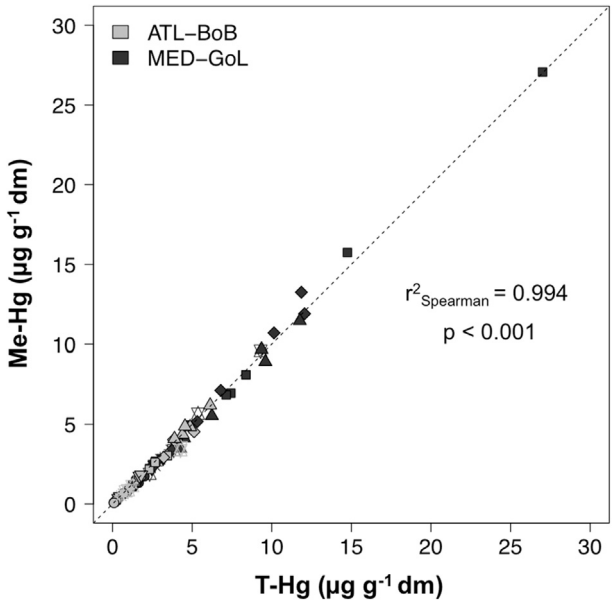

Fig. 3. Relationship between muscle methyl-Hg (Me-Hg) and total Hg (T-Hg) concentrations for the subsample of individuals analysed for Me-Hg concentrations (n = 75), and for the seven species analysed in the NW Mediterranean – Gulf of Lion (MED – GoL) and in the NE Atlantic – Bay of Biscay (ATL – BoB). The correspondence line 1:1 is indicated, as well as the results of the non-parametric Spearman correlation coefficient test. Symbols used for species are the same than in Fig. 2c.

of the species examined, although characteristic offshore species of the shelf-edge in both systems, revealed the significant differences between the two systems in terms of trophic status and consequently in terms of sources of organic matter and primary producers sustaining the respective food webs. Unfortunately, no dedicated sampling of plankton could be carried out for the present study to get the $\delta^{15}N$ values (and Hg concentrations) of the planktonic compartment, which could be directly related to those of the medium to high-trophic level consumers here of concern. However, the $\delta^{15}N$ values of the consumers considered here clearly showed the more oligotrophic character of the Mediterranean waters in general. Indeed, the fixation of atmospheric N_2 by diazotrophic organisms (e.g., cyanobacteria) in offshore and oligotrophic waters is well documented to lower the $\delta^{15}N$ values of the residual NO_3^- pool available for primary producers in these areas (see review by Montoya (2007). Diazotrophic organisms were demonstrated to be preponderant in the GoL (Kerhervé et al., 2001; Le Moal and Biegala, 2009; Liénart et al., 2017), thus resulting in low $\delta^{15}N$ values in consumers at all trophic levels (especially when compared to their counterparts in the BoB waters, for instance), from mesozooplankton (Chouvelon et al., 2012, 2014b; Espinasse et al., 2014; Bănaru et al., 2014) and suspension-feeders like bivalves (Cresson et al., 2016; Briant et al., 2018) to deep-sea fish and crustaceans (present study; Cresson et al., 2014b). Finally, the results of the present study highlighted the higher primary productivity of the BoB offshore waters (i.e. higher $\delta^{15}N$ values measured in individuals from this ecosystem) compared to offshore GoL waters. In the BoB, in the Cap-Ferret canyon's head area (i.e. offshore area) for instance, the annual primary productivity was effectively estimated to be 145–170 gC/m^2 (Laborde et al., 1999), while it was estimated to be 78–142 gC/m^2 in the GoL (Lefevre et al., 1997).

The significant difference of sizes observed for most of the species considered here, despite comparable sampling methodology and effort, were also indicative of a difference in individual size distribution between the two systems with contrasted trophic status. This difference is likely due to the lower growth rate of Mediterranean organisms compared their Atlantic counterparts, like observed for the hake *M. merluccius*, for instance (De Pontual et al., 2006; Mellon-Duval et al., 2009). As such, for similar-sized individuals, organisms from the Mediterranean are surely much older than their Atlantic counterparts. In the previous study by Cossa et al. (2012), this partly explained the higher concentrations of Hg observed in Mediterranean hakes compared to Atlantic ones due to age-related bioaccumulation of the metal, in addition to the fact that Hg burden is also more bio-diluted in larger organisms.

4.2. Me-Hg/T-Hg ratios, bioaccumulation and biomagnification of Hg

The relatively high percentage of Me-Hg measured (i.e. >85% on

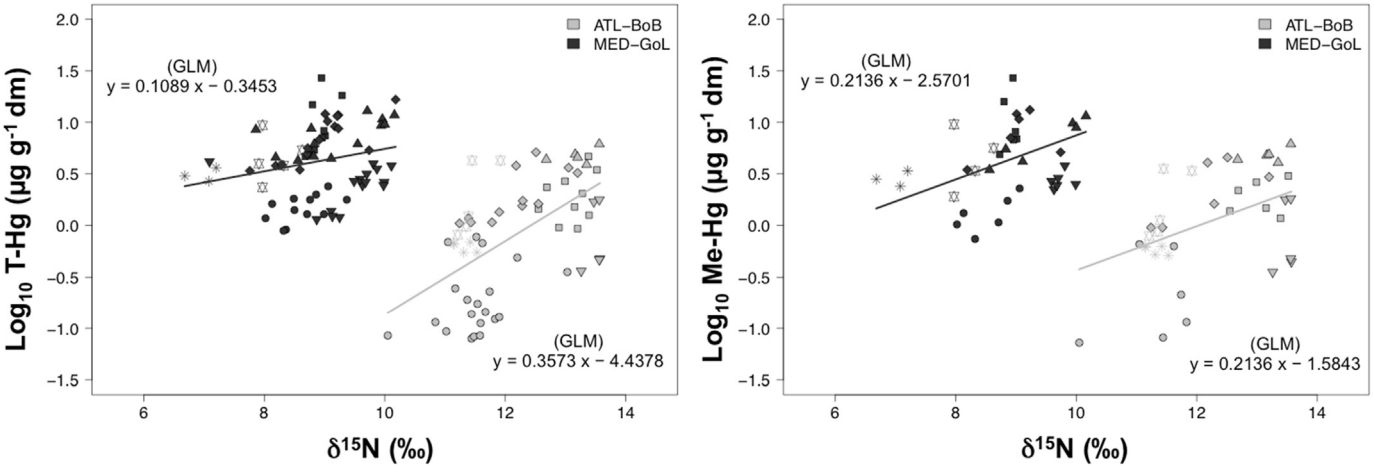

Fig. 4. Relationships between individual log-transformed total Hg (T-Hg) concentrations (left panel, N = 134) or methyl-Hg (Me-Hg) concentrations (right panel, n = 75), and $\delta^{15}N$ values (as a proxy of the trophic level within a given systems), for the seven species analysed in the NW Mediterranean – Gulf of Lion (MED – GoL) and in the NE Atlantic – Bay of Biscay (ATL – BoB). The symbology used for each species is the same as in Fig. 2c. Results from the GLMs (lines) are plotted on observed log-transformed data. When the interaction term was not significant (i.e. for MeHg, the model assumes that the form of the relationship is not different between the ecosystems), lines are parallel (i.e. same slope, although different intercept). This can also be interpreted (i.e. slope of the lines) as the biomagnification power of T-Hg or Me-Hg within each ecosystem considered (as performed by Cossa et al., 2012; for Me-Hg). Finally, the equations of lines (derived from model outputs; Table 2) are indicated.

average for all species and in both ecosystems) was expected, since muscle was the tissue examined, and since the trophic position of the species considered was quite elevated. Indeed, this percentage has long been considered virtually equal to 100% in the muscle of high-trophic level consumers such as fish and cephalopod molluscs (e.g., Bloom, 1992), due to the high affinity of Hg for muscular proteins' sulfhydryl groups, although it generally varies between 60 and 90% depending on species and on their trophic level (e.g., Bustamante et al., 2006; Kehrig et al., 2010; Cossa et al., 2012). It may be also elevated in some crustacean species, but it is clearly lower (generally largely <50%) in low-trophic level consumers such as bivalve molluscs (e.g., Cossa et al., 2012; Briant et al., 2017). Opposite to the other parameters regarded (i.e. individual sizes, $\delta^{13}C$ and $\delta^{15}N$ values, raw T-Hg and Me-Hg concentrations), the percentage of Me-Hg in the muscle of the selected organisms generally did not differ a lot between the BoB and the GoL organisms. These results are in agreement with those of Cossa et al. (2012) that observed no significant differences in the percentage of Me-Hg in Mediterranean vs. Atlantic hakes. The fact that only medium-to high-trophic levels were considered here may partly explain this absence of difference in the Me-Hg/T-Hg ratios between organisms from the two systems. Indeed, when considering organisms at the top of food webs, potential differences that could exist in the Me-Hg/T-Hg proportions at the basal levels (in seawater, and/or in low-trophic level organisms) may no longer be visible.

In fact, water Me-Hg concentrations were shown to be comparable between the BoB and the GoL shelf-edge ecosystems (Cossa et al., 2012). However, the BoB has probably a deeper thermohalocline than the GoL and consequently a deeper organic matter regeneration zone, where Hg is likely methylated (Heimbürger et al., 2010; Blum et al., 2013; Cossa et al., 2017). As the species considered here are endemic of the shelf-edge and heads of canyons, they may be therefore more exposed to Me-Hg in Mediterranean waters than in Atlantic ones (through the higher content of Me-Hg in the lower trophic levels themselves). This hypothesis is also supported by the lower percentage of Me-Hg measured for *M. poutassou* in the GoL in particular, relative to the other fish species examined. Indeed, the shallower foraging zone of this species was previously proposed as an explanation of its lower T-Hg burdens (Cresson et al., 2014b), but this may also explain the lower proportion of Me-Hg in the muscle of this species, in addition to its lower trophic level in comparison with other species.

The significant relationships between log-transformed Hg concentrations and $\delta^{15}N$ values were in agreement with the species-dependent patterns previously observed (i.e. increasing concentrations with increasing trophic levels), and were thus also consistent with the well-documented property of Hg to biomagnify in food webs (Boening, 2000). Furthermore, the equations of these relationships allow to compare the BP of this metal across food webs, using the slope of the relationships as a measure of the biomagnification rate, and the intercept as the baseline value for primary producers in the different systems (Chen et al., 2008; Cossa et al., 2012; Lavoie et al., 2013). Then, the biomagnification power (BP) of Hg may be expected to differ between systems when the $\delta^{15}N$ values of plankton (i.e. low trophic levels) differ between systems, and/or when Hg concentrations in plankton differ between systems. Besides, variations in the $\delta^{15}N$ values or Hg concentrations of plankton are linked, at least part, to the composition of plankton (some planktonic species being able to fix atmospheric nitrogen with peculiar N signature, for instance; Kerhervé et al., 2001). As a consequence, the BP may be expected to differ between systems with different trophic status, i.e. between systems with different planktonic communities at the base of food webs.

Here, higher intercepts were found for the Mediterranean ecosystem, indicating a probably higher Hg burden from the lowest trophic levels in the GoL compared to BoB (i.e. lower "bio-dilution" effect in the GoL's low trophic levels than in those from the BoB). However, the intercept is intimately linked to the slope, and such direct link with Hg baseline values should be done with caution when the lowest trophic levels (plankton) are not available (Borgå et al., 2012), as it is the case for the present study. Biomagnification slopes (i.e. BPs), were previously reported to range between 0.09 and 0.22 for T-Hg (average 0.20 ± 0.10) and between 0.14 and 0.26 for Me-Hg (average 0.22 ± 0.09) in marine ecosystems (Lavoie et al., 2013). When considering Me-Hg concentrations, the BPs measured here (0.21 for both BoB and GoL ecosystems, not significantly different) are therefore within this range of values. Alternatively, when calculated from T-Hg concentrations, the BPs significantly differed between the BoB (0.35) and the GoL (0.11), with a lower BP estimated for the Mediterranean system. This differs from the results of Cossa et al. (2012), for instance, which found a higher BP in the hake food web from the GoL (Mediterranean) than in those from the BoB (Atlantic). The consideration of different food webs and organisms may be at the origin of the discrepancy between our study and those of Cossa et al. (2012) (i.e. consideration of the neritic hake food web by Cossa et al., 2012, including lower trophic levels, vs. shelf-edge/offshore organisms and medium-to high-trophic levels only here). Moreover, the lower BP found here for the Mediterranean ecosystem may favour the hypothesis of potentially higher basal Hg levels (i.e. in plankton) than in Atlantic for the shelf-edge food webs considered here (as suggested by the different intercept as well), although the biomagnification potential of Hg may be then not especially higher in the rest of the food web (as suggested by similar BPs when considering the bioavailable and bioaccumulable form Me-Hg). Finally, if our estimated BPs probably remain comparable at the scale of our study between GoL and BoB, with the same species considered in both systems, they are not necessarily comparable to other studies that would include other (non-fish) species and especially lower trophic levels (e.g., Signa et al., 2017).

Overall, the present study thus showed that the higher Hg bioaccumulation rate by Mediterranean organisms is likely linked to the oligotrophic character of waters and associated lower productivity in this system. Indeed, compared to mesotrophic environments, oligotrophic conditions are likely associated with: i) increased formation of the bioavailable Me-Hg at the depths of organic matter regeneration (i.e. where the shelf-edge/offshore species considered here likely live), due to greater proportions of slowly sinking pico- and nanophytoplanctonic cells that are readily degraded by bacteria there (Cossa et al., 2009; Heimbürger et al., 2010); ii) lower bio-dilution of Hg from the lower trophic levels (i.e. phytoplankton; Pickhardt et al., 2002), with the presence of smaller and less abundant cells that are then also much more consumed – including their high Hg burden – by higher trophic level organisms; iii) lower growth rate of consumer organisms and consequently, lower bio-dilution of Hg burden in their tissues as well and high Hg retention with the age of organisms, the elimination rate of the bioaccumulated Hg being very low (Wang and Wong, 2003; Maulvault et al., 2016).

Finally, the potential impact of temperature differences between GoL and BoB on Hg bioaccumulation is also interesting to address in the context of global change. Higher temperatures are likely to enhance bacterial activity and consequently Me-Hg formation, which in our study case may also partly explain the higher Hg bioaccumulation observed in Mediterranean organisms. Indeed in the GoL, the sea surface temperatures were shown to vary between 14 and 20 °C and to remain relatively constant around 13 °C below 100–200 m depth (Conan et al., 1998), while in the BoB, the sea surface temperatures are slightly colder. They vary between 11 and

20 °C (for the southern part, less in the northern one) and remain below 12 °C in depth (Koutsikopoulos and Le Cann, 1996). In addition, higher temperatures such as it is likely the case for Mediterranean waters were proved to promote Me-Hg bioaccumulation and to hamper its elimination in fish tissues, for instance (Maulvault et al., 2016), which is also in favour of higher potential for Hg bioaccumulation in Mediterranean organisms.

5. Conclusion and perspectives

The effect of ecosystems' characteristics on Hg bioaccumulation and biomagnification was already demonstrated in freshwater ecosystems (e.g., lakes) through comparative studies (e.g., Kidd et al., 2012). Such ecosystem-comparative or meta-analysis studies remained rare for marine ecosystems. They were therefore recently encouraged to be done for generating new testable hypotheses concerning Hg bioaccumulation and biomagnification (Lavoie et al., 2013).

Benefiting from archived samples from the GoL and the BoB shelf-edge/offshore ecosystems, our study showed the strong influence of the trophic status and/or functioning of systems on Hg bioaccumulation in marine medium-to high-trophic level organisms, which are generally important commercial species. It also reinforced and confirmed the hypothesis made by Cossa and Coquery (2005) that biological processes dominate the geochemical ones in explaining the "Mediterranean mercury anomaly". Furthering some previous studies focusing on the coastal or neritic species that are red mullets or hakes (Cossa et al., 2012; Cresson et al., 2014a, 2015), the present marine ecosystem-comparative study demonstrated the crucial role of oligotrophy and associated lower productivity on Hg bioaccumulation, at both multi-species and multi-systems scales.

As the processes of bio-dilution and low elimination rates may also apply to other metals, we propose that the bioaccumulation of other metals may be also determined by the trophic status of the considered ecosystems in a non-negligible part. Thus, it would be interesting to examine other trace metals on the samples used in the present study, to investigate whether oligotrophy may also influence their bioavailability, transfer and bioaccumulation, and whether the patterns observed for Hg are also applicable or not for other trace elements (e.g., species-dependant patterns kept or not between the systems). Also, investigating the potential differences in the energy content of prey species/lower trophic levels between systems (i.e. expected to be affected by oligotrophy in Mediterranean waters, for instance) may be interesting. This would enable to more globally apprehend the mechanisms involved in the transfer of contaminants (especially the lipophilic ones) to higher trophic levels, and their transfer in food webs in general. Finally, with regard to the high significance of the trophic status of systems on Hg bioaccumulation shown in the present study, further work on very low trophic levels (i.e. plankton) would be also highly relevant.

Acknowledgments

Authors are particularly grateful to the "Agence de l'Eau Rhône Méditerranée Corse" (AERMC) for financial support of the Mediterranean analyses and of the associated RETROMED project, as well as to the "Conseil Régional de Poitou-Charentes" and the "Contrat de projet Etat-Région" (CPER 2013) that supported the analyses relative to the Bay of Biscay area during the PhD of T. Chouvelon. We are grateful to C. Churlaud from the "Plateforme Analyses Elémentaires" of LIENSs for her assistance during Hg analyses and to G. Guillou, P. Richard and B. Lebreton from the "Plateforme de Spectrométrie Isotopique" of the LIENSs laboratory for their assistance during SIA analyses. Thanks are also due to M.C. Fabri for its implication in the RETROMED project, and to F. Chavanon and C. Ravel for their help during sampling and laboratory work. Finally, authors want to thank the chief scientists and the crew of the R/V "Thalassa" and "L'Europe" for facilitating the sampling during EVHOE 2008 (https://doi.org/10.17600/8040120) and MEDITS 2012 (https://doi.org/10.17600/12060100) surveys.

Appendix A. Supplementary data

Supplementary data related to this article can be found at https://doi.org/10.1016/j.envpol.2017.11.015.

References

Aston, S.R., Fowler, S.W., 1985. Mercury in the open Mediterranean: evidence of contamination? Sci. Total Environ. 43, 13–26.
Azemard, S., Vassileva, E., 2015. Determination of methylmercury in marine biota samples with advanced mercury analyser: method validation. Food Chem. 176, 367–375.
Bănaru, D., Carlotti, F., Barani, A., Grégori, G., Neffati, N., Harmelin-Vivien, M., 2014. Seasonal variation of stable isotope ratios of size-fractionated zooplankton in the Bay of Marseille (NW Mediterranean Sea). J. Plankton Res. 36, 145–156.
Biddanda, B., Ogdahl, M., Cotner, J., 2001. Dominance of bacterial metabolism in oligotrophic relative to eutrophic waters. Limnol. Oceanogr. 46, 730–739.
Bloom, N.S., 1992. On the chemical form of mercury in edible fish and marine invertebrate tissue. Can. J. Fish. Aquat. Sci. 49, 1010–1017.
Blum, J.D., popp, B.N., Drazen, J.C., Choy, C.A., Johnson, M.W., 2013. Methylmercury production below the mixed layer in the north pacific ocean. Nat. Geosci. 6, 879–884.
Boening, D., 2000. Ecological effects, transport, and fate of mercury: a general review. Chemosphere 40, 1335–1351.
Borgå, K., Kidd, K.A., Muir, D.C.G., Berglund, O., Conder, J.M., Gobas, F.A.P.C., Kucklick, J., Malm, O., Powell, D.E., 2012. Trophic magnification factors: considerations of ecology, ecosystems, and study design. Integr. Envrion Assess. Manag. 8, 64–84.
Briant, N., Chouvelon, T., Martinez, L., Brach-Papa, C., Chiffoleau, J.F., Savoye, N., Sonke, J., Knoery, J., 2017. Spatial and temporal distribution of mercury and methylmercury in bivalves from the French coastline. Mar. Pollut. Bull. 114, 1096–1102.
Briant, N., Savoye, N., Chouvelon, T., David, V., Rodriguez, S., Charlier, K., Sonke, J., Chiffoleau, J.F., Brach-Papa, C., Knoery, J., 2018. Carbon and nitrogen elemental and isotopic ratios of filter-feeding bivalves along the French coasts: an assessment of specific, geographic, seasonal and multi-decadal variations. Sci. Tot Environ. 613–614, 196–207.
Bustamante, P., Lahaye, V., Durnez, C., Churlaud, C., Caurant, F., 2006. Total and organic Hg concentrations in cephalopods from the North Eastern Atlantic waters: influence of geographical origin and feeding ecology. Sci. Total Environ. 368, 585–596.
Chen, C., Amirbahman, A., Fisher, N., Harding, G., Lamborg, C., Nacci, D., Taylor, D., 2008. Methylmercury in marine ecosytems: spatial patterns and processes of production, bioaccumulation, and biomagnification. EcoHealth 5, 399–408.
Chen, C.Y., Folt, C.L., 2005. High plankton densities reduce mercury biomagnification. Environ. Sci. Technol. 39, 115–121.
Chouvelon, T., Spitz, J., Caurant, F., Mèndez-Fernandez, P., Autier, J., Lassus-Débat, A., Bustamante, P., 2012. Enhanced bioaccumulation of mercury in deep-sea fauna from the Bay of Biscay (North-East Atlantic) in relation to trophic positions identfied by analysis of carbon and nitrogen stable isotopes. Deep-Sea Res. I 65, 113–124.
Chouvelon, T., Caurant, F., Cherel, Y., Spitz, J., Simon-Bouhet, B., Bustamante, P., 2014a. Species- and size-related patterns in stable isotopes and mercury concentrations in fish help refine marine ecosystem indicators and provide evidence for distinct management units for hake in the Northeast Atlantic. ICES J. Mar. Sci. 75, 1073–1087.
Chouvelon, T., Chappuis, A., Bustamante, P., Lefebvre, S., Mornet, F., Guillou, G., Violamer, L., Dupuy, C., 2014b. Trophic ecology of European sardine *Sardina pilchardus* and European anchovy *Engraulis encrasicolus* in the Bay of Biscay (North-East Atlantic) inferred from $\delta^{13}C$ and $\delta^{15}N$ values of fish and identified mesozooplanktonic organisms. J. Sea Res. 85, 277–291.
Chouvelon, T., Brach-Papa, C., Auger, D., Bodin, N., Bruzac, S., Crochet, S., Degroote, M., Hollanda, S.J., Hubert, C., Knoery, J., Munschy, C., Puech, A., Rozuel, E., Thomas, B., West, W., Bourjea, J., Nikolic, N., 2017. Chemical contaminants (trace metals, persistent organic pollutants) in albacore tuna from western Indian and south-eastern Atlantic Oceans: Trophic influence and potential as tracers of populations. Science of the Total Environment 596-597, 481–495.
Choy, C.A., Popp, B.N., Kaneko, J.J., Drazen, J.C., 2009. The influence of depth on mercury levels in pelagic fishes and their prey. Proc. Natl. Acad. Sci. U. S. A. 106, 13865–13869.
Cossa, D., Averty, B., Pirrone, N., 2009. The origin of methylmercury in open Mediterranean waters. Limnol. Oceanogr. 54, 837–844.

Conan, P., Pujo-Pay, M., Raimbault, P., Leveau, M., 1998. Variabilité hydrologique et biologique du golfe du Lion. II. Productivité sur le bord interne du courant. Oceanol. Acta 21, 767–782.
Cossa, D., Coquery, M., 2005. The Mediterranean mercury anomaly, a geochemical or a biological issue. In: Saliot, A. (Ed.), The Mediterranean Sea. Handbook of Environmental Chemistry, vol. 5. Springer Berlin Heidelberg, pp. 177–208.
Cossa, D., Harmelin-Vivien, M., Mellon-Duval, C., Loizeau, V., Averty, B., Crochet, S., Chou, L., Cadiou, J.F., 2012. Influences of bioavailability, trophic position, and growth on methylmercury in hakes (*Merluccius merluccius*) from Northwestern Mediterranean and Northeastern Atlantic. Environ. Sci. Technol. 46, 4885–4893.
Cossa, D., Durrieu de Madron, X., Schäfer, J., Lanceleur, L., Guédron, S., Buscail, R., Thomas, B., Castelle, S., Naudin, J.J., 2017. The open sea as the main source of methylmercury in the water column of the Gulf of Lions (Northwestern Mediterranean margin). Geochim. Cosmochim. Acta 199, 222–237.
Cresson, P., Bouchoucha, M., Miralles, F., Elleboode, R., Mahé, K., Marusczak, N., Thebault, H., Cossa, D., 2014a. Are red mullet efficient as bio-indicators of mercury contamination? A case study from the French Mediterranean? Mar. Pollut. Bull. 91, 191–199.
Cresson, P., Fabri, M.C., Bouchoucha, M., Brach Papa, C., Chavanon, F., Jadaud, A., Knoery, J., Miralles, F., Cossa, D., 2014b. Mercury in organisms from the Northwestern Mediterranean slope: importance of food sources. Sci. Tot Environ. 497–498, 229–238.
Cresson, P., Bouchoucha, M., Morat, F., Miralles, F., Chavanon, F., Loizeau, V., Cossa, D., 2015. A multitracer approach to assess the spatial contamination pattern of hake (*Merluccius merluccius*) in the French Mediterranean. Sci. Tot Environ. 532, 184–194.
Cresson, P., Ruitton, S., Harmelin-Vivien, M., 2016. Feeding strategies of co-occurring suspension feeders in an oligotrophic environment. Food Webs 6, 19–28.
De Pontual, H., Groison, A.L., Pineiro, C., Bertignac, M., 2006. Evidence of underestimation of European hake growth in the Bay of Biscay, and its relationship with bias in the agreed method of age estimation. ICES J. Mar. Sci. 63, 1674–1681.
Espinasse, B., Harmelin-Vivien, M., Tiano, M., Guilloux, L., Carlotti, F., 2014. Patterns of variations in C and N stable isotope ratios in size-fractionated zooplankton in the Gulf of Lion, NW Mediterranean Sea. J. Plankton Res. 36, 1204–1215.
Fitzgerald, W.F., Lamborg, C.H., Hammerschmidt, C.R., 2007. Marine biogeochemical cycling of mercury. Chem. Rev. 107, 641–662.
Goutte, A., Bustamante, P., Barbraud, C., Delord, K., Weimeskirch, H., Chastel, O., 2014. Demographic responses to mercury exposure in two closely-related Antarctic top predators. Ecology 95, 1075–1086.
Harmelin-Vivien, M., Cossa, D., Crochet, S., Banaru, D., Letourneur, Y., Mellon-Duval, C., 2009. Difference of mercury bioaccumulation in red mullets from the north-western Mediterranean and Black seas. Mar. Pollut. Bull. 58, 679–685.
Heimbürger, L.E., Cossa, D., Marty, J.C., Migon, C., Averty, B., Dufour, A., Ras, J., 2010. Methyl mercury distributions in relation to the presence of nano- and picophytoplankton in an oceanic water column (Ligurian Sea, North-western Mediterranean). Geochim. Cosmochim. Acta 74, 5549–5559.
Kehrig, H.A., Seixas, T.G., Baêta, A.P., Malm, O., Moreira, I., 2010. Inorganic and methylmercury: do they transfer along a tropical coastal food web? Mar. Pollut. Bull. 60, 2350–2356.
Kerhervé, P., Minagawa, M., Heussner, S., Monaco, A., 2001. Stable isotopes ($^{13}C/^{12}C$ and $^{15}N/^{14}N$) in settling organic matter of the northwestern Mediterranean Sea: biogeochemical implications. Oceanol. Acta 24, S77–S85.
Kidd, K.A., Muir, D.C.G., Evans, M.S., Wang, X., Whittle, M., Swanson, H.K., Johnston, T., Guildford, S., 2012. Biomagnification of mercury through lake trout (*Salvelinus namaycush*) food webs of lakes with different physical, chemical and biological characteristics. Sci. Tot Environ. 438, 135–143.
Koutsikopoulos, C., Le Cann, B., 1996. Physical processes and hydrological structures related to the Bay of Biscay anchovy. Sci. Mar. 60, 9–19.
Laborde, P., Urrutia, J., Valencia, V., 1999. Seasonal variability of primary production in the Cap-Ferret Canyon area (Bay of Biscay) during the ECOFER cruises. Deep-Sea Res. II 2057–2079.
Lavoie, R.A., Jardine, T.D., Chumchal, M.M., Kidd, K.A., Campbell, L.M., 2013. Biomagnification of mercury in aquatic food webs: a worldwide meta-analysis. Environ. Sci. Technol. 47, 13385–13394.
Le Faucheur, S., Campbell, P.G.C., Fortin, C., Slaveykova, V.I., 2014. Interactions between mercury and phytoplankton: speciation, bioavailability, and internal handling. Environ. Toxicol. Chem. 33, 1211–1224.
Lefevre, D., Minas, H.J., Minas, M., Robinson, C., Williams, P.J., Le, B., Woodward, E.M.S., 1997. Review of gross community production, primary production, net community production and dark community respiration in the Gulf of Lions. Deep-Sea Res. II 44, 801–832.
Le Moal, M., Biegala, I.C., 2009. Diazotrophic unicellular cyanobacteria in the northwestern Mediterranean Sea: a seasonal cycle. Limnol. Oceanogr. 54, 844–855.
Liénart, C., Savoye, N., Bozec, Y., Breton, E., Conan, P., David, V., Feunteun, E., Grnagéré, K., Kerhervé, P., Lebreton, B., Lefebvre, S., et al., 2017. Dynamics of particulate organic matter composition in coastal systems: a spatio-temporal study at multi-systems scale. Prog. Oceanogr. 156, 221–239.
Longhurst, A., 1998. Ecological Geography of the Sea, vol. 1. Academic Press, London, p. 560.
Longhurst, A., 2007. Ecological Geography of the Sea, vol. 2. Academic Press, London, p. 398.
Mason, R.P., Rolfhus, K.R., Fitzgerald, W.F., 1995. Methylated and elemental mercury cycling in surface and deep ocean waters of the North Atlantic. Wat Air Soil Pollut. 80, 665–677.
Maulvault, A.L., Custódio, A., Anacleto, P., Repolho, T., Pousão, P., Nunes, M.L., Diniz, M., Rosa, R., Marques, A., 2016. Bioaccumulation and elimination of mercury in juvenile seabass (*Dicentrarchus labrax*) in a warmer environment. Environ. Res. 149, 77–85.
Mellon-Duval, C., de Pontual, H., Métral, L., Quemener, L., 2009. Growth of European hake (*Merluccius merluccius*) in the Gulf of Lions based on conventional tagging. ICES J. Mar. Sci. 67, 62–70.
Mèndez-Fernandez, P., Pierce, G.J., Bustamante, P., Chouvelon, T., Ferreira, M., González, A.F., López, A., Read, F., Santos, M.B., Spitz, J., Vingada, J.V., Caurant, F., 2013. Ecological niche segregation among five toothed whale species off the NW Iberian Peninsula using ecological tracers as multi-approach. Mar. Biol. 160, 2825–2840.
Monteiro, L.R., Costa, V., Furness, R.W., Santos, R.S., 1996. Mercury concentrations in prey fish indicate enhanced bioaccumulation in mesopelagic environments. Mar. Ecol. Prog. Ser. 141, 21–25.
Monteiro, L.R., Lopes, H.D., 1990. Mercury content of swordfish, *Xiphias gladius*, in relation to length, weight, age, and sex. Mar. Pollut. Bull. 21, 293–296.
Montoya, J.P., 2007. Natural abundance of ^{15}N in marine planktonic ecosystems. In: Michener, R., Lajtha, K. (Eds.), Stable Isotopes in Ecology and Environmental Science. Blackwell Publishing, pp. 76–201.
Pickhardt, P.C., Folt, C.L., Chen, C.Y., Klaue, B., Blum, J.D., 2002. Algal blooms reduce the uptake of toxic methylmercury in freshwater food webs. Proc. Natl. Acad. Sci. U. S. A. 99, 4419–4423.
Pierce, G.J., Stowasser, G., Hastie, L.C., Bustamante, P., 2008. Geographic, seasonal and ontogenetic variation in cadmium and mercury concentrations in squid (Cephalopoda: teuthoidea) from UK waters. Ecotoxicol. Environ. Saf. 70, 422–432.
Reygondeau, G., longhurst, A., Martinez, E., Beaugrand, G., Antoine, D., Maury, O., 2013. Dynamic biogeochemical provinces in the global ocean. Glob. Biogeochem. Cy 27, 1–13.
Signa, G., Mazzola, A., Tramati, C.D., Vizzini, S., 2017. Diet and habitat use influence Hg and Cd transfer to fish and consequent biomagnification in a highly contaminated area: augusta Bay (Mediterranean Sea). Environ. Pollut. 230, 394–404.
Simoneau, M., Lucotte, M., Garceau, S., Laliberte, D., 2005. Fish growth rates modulate mercury concentrations in walleye (*Sander vitreus*) from eastern Canadian lakes. Environ. Res. 98, 73–82.
Tan, S.W., Meiller, J.C., Mahaffey, K.R., 2009. The endocrine effects of mercury in humans and wildlife. Crit. Rev. Toxicol. 39, 228–269.
Trudel, M., Rasmussen, J.B., 2006. Bioenergetics and mercury dynamics in fish: a modelling perspective. Can. J. Fish. Aquat. Sci. 63, 1890–1902.
US EPA, 2011. Exposure Factors Handbook 2011 Edition. Final Report. US Environmental Protection Agency, Washington, DC. EPA/600/R-09/052F.
Wang, W.X., Wong, R.S.K., 2003. Bioaccumulation kinetics and exposure pathways of inorganic mercury and methylmercury in a marine fish, the sweetlips *Plectorhinchus gibbosus*. Mar. Ecol. Prog. Ser. 257, 257–268.
Ward, D.M., Nislow, K.H., Chen, C.Y., Folt, C.L., 2010. Reduced trace element concentrations in fast-growing juvenile Atlantic salmon in natural streams. Environ. Sci. Technol. 44, 3245–3251.
Zuur, A.F., Ieno, E.N., Smith, G.M., 2007. Analysing Ecological Data. Springer, New York (USA).