Environmental Science & Technology

Article

pubs.acs.org/est

Influences of Bioavailability, Trophic Position, and Growth on Methylmercury in Hakes (*Merluccius merluccius*) from Northwestern Mediterranean and Northeastern Atlantic

D. Cossa,*,† M. Harmelin-Vivien,‡ C. Mellon-Duval,§ V. Loizeau,∥ B. Averty,⊥ S. Crochet,⊥ L. Chou,# and J.-F. Cadiou†

†Ifremer, Centre de Méditerranée, BP 330, F-83507 La Seyne-sur-Mer, France

‡Aix-Marseille Université, Institut Méditerranéen d'Océanologie, UMR CNRS 7294, 13007 Marseille, France

§Ifremer, Centre de Recherche Halieutique Méditerranéenne et Tropicale, BP 171, F-34203 Sète, France

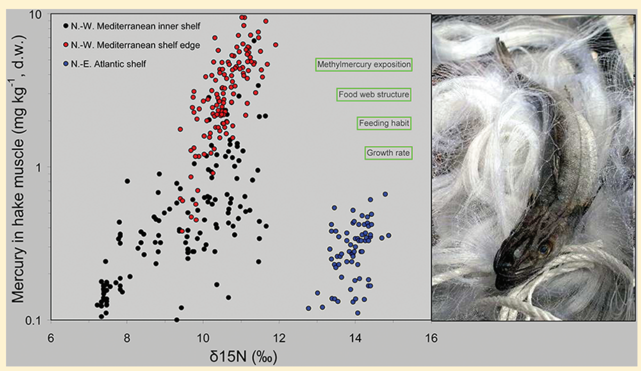
∥Ifremer, Centre de Brest, BP 90, F-29280 Plouzané, France

⊥Ifremer, Centre Atlantique, BP 21105, F-44311 Nantes Cedex 03, France

#Chemical Oceanographic Laboratory, Université Libre de Bruxelles, B-1050 Brussels, Belgium

Supporting Information

ABSTRACT: Methylmercury (MeHg) determinations in hake, its food-chain, and the surrounding waters and sediments allowed us to show that the higher length or age normalized mercury concentrations of Northwestern Mediterranean (Gulf of Lions: GoL) muscle hakes compared to its Northeastern Atlantic (Bay of Biscay: BoB) counterpart are due to both biotic and abiotic differences between their ecosystems. Bioenergetic modeling reveals that the slower growth rate of Mediterranean hake favors the MeHg bioaccumulation in the fish muscle and explains most of the difference between GOL and BoB hake populations. In addition, the waters of the Mediterranean hake habitat favor a higher MeHg exposition, due to the upper position of the thermohalocline, where MeHg is formed. Furthermore, we show that, within the Mediterranean hake population, a major increase in the biomagnification power (the slope of the relationships between logMeHg and $\delta^{15}N$), from 0.36 up to 1.12, occurs when individuals enter adulthood, resulting from the combined effects of lowering growth rate and change in feeding habits. Finally, $\delta^{15}N$ normalized Hg concentrations indicate that the highest Hg concentrations are for hake from the shelf edge and the lowest are for hake from the Rhône prodelta area, suggesting a lower Hg bioavailability in inshore environments, consistent with MeHg distributions in water, sediment, and preys.



INTRODUCTION

The high concentrations of mercury in marine top fish predators is a long-standing concern, which results from the biomagnification of the methylmercury molecule (MeHg) through trophic webs.[1−3] Methylmercury is easily taken up by algae, with a huge bioconcentration factor (>10^5),[4−6] and efficiently absorbed via the digestive tracts of predators.[7] Most of the MeHg in fish tissue is covalently bound to protein sulfhydryl groups,[8,9] and shows a very slow elimination rate.[10] Consequently, in a given fish species, MeHg concentrations tend to increase with age with a speed that depends on their growth rate. This ends with MeHg representing virtually the total amount of mercury in the muscle tissue of the greatest predators.[11,12] The biochemical integration of MeHg in dynamic tissue results in "growth dilution", as demonstrated for plankton[13] and fish.[14] In summary, the bioavailability of methylmercury for plankton, primary productivity, the structure of the food chain, the growth rate, and the age of the predators need to be taken into account to understand the MeHg concentration distribution and dynamic in predator fishes from various marine environments.

For decades, numerous studies have pointed out higher total mercury (Hg_T) concentrations in Mediterranean marine organisms than in the same species living in the adjacent North Atlantic[15−18] or in the Black Sea.[19] These discrepancies in organism Hg concentration in different environments were particularly noticeable for top predators, such as tuna or mammals.[20,21] Aston and Fowler[22] reviewed the main hypotheses to explain these observations. They mentioned the lack of qualified data for inorganic and organic mercury in waters and various components of the ecosystem, and

Received: December 6, 2011
Revised: April 6, 2012
Accepted: April 6, 2012
Published: April 6, 2012

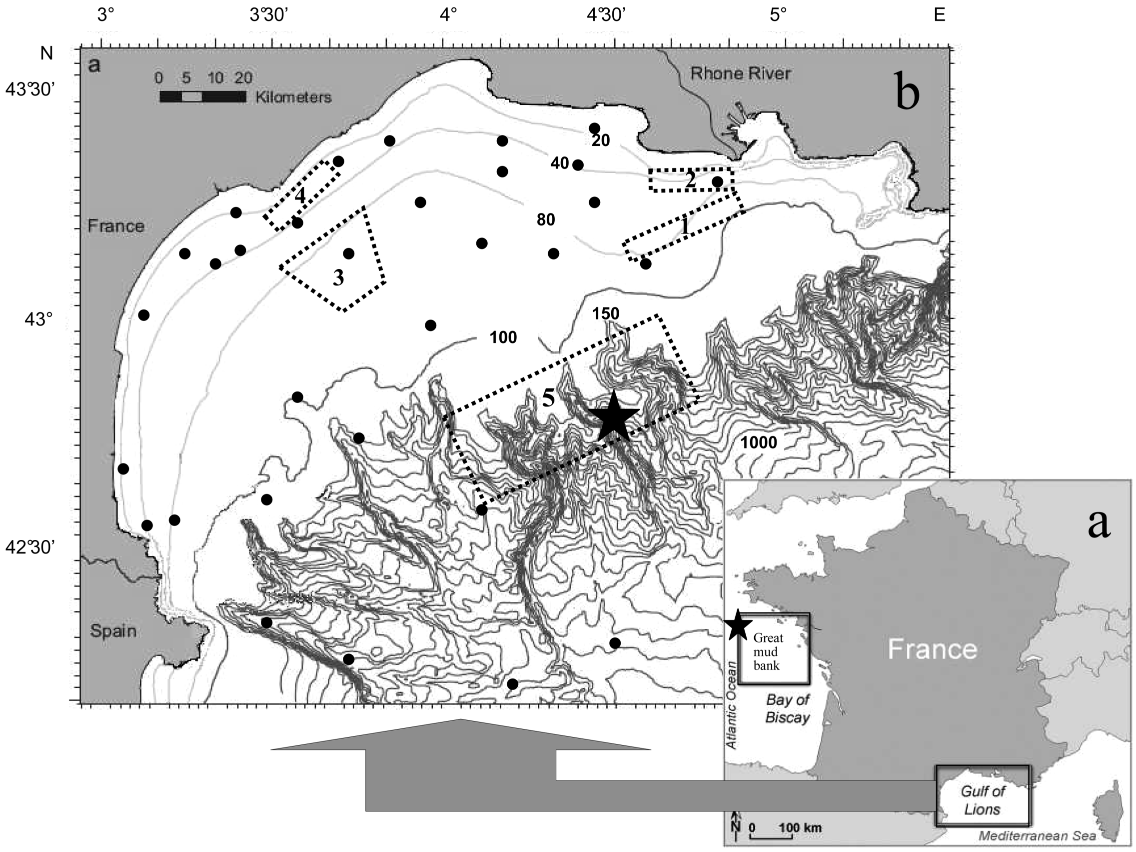


Figure 1. Sampling sites in Gulf of Lions (GoL) and Bay of Biscay (BoB). (a) GoL and BoB shelves areas; trawling areas are within the Great mud bank for BoB (a) and within the five dot-lined polygons (b) for GoL (sectors 1 and 2 are located in the Rhône prodelta area); black dots indicate the sediment sites (b) and star points indicate the areas of the water column sampling stations (a and b).

ecological factors inherent to the Mediterranean Sea (growth conditions, food webs, etc.), which may control the biomagnification processes in this particular environment. In the last ten years new data on mercury speciation in the atmosphere, water, and sediments have permitted a better understanding of the mercury cycle in the Mediterranean basins,[23−27] but no definitive answer has really been given to the issue of the particularly high Hg biomagnification in Mediterranean fish. Recently, low biological production at the base of the trophic web in the Mediterranean (generating a low dilution effect) was argued as a reason for the higher Hg levels found in the muscle of red mullet from the Gulf of Lions compared to those from the Black Sea,[19] since the methylmercury level in the water (a good proxy for Hg avalability for plankton) has not been proved to be different between these two environments.

The present paper intends to address the characteristics and specificities of the mercury biomagnification in the European hake from the Gulf of Lions (GoL) in the Northwestern Mediterranean, and comparing these with results obtained from the Bay of Biscay (BoB) in the Northeastern Atlantic. Four specific questions will be addressed: what is the mercury bioavailability in the studied environments?; what are the effects of trophic position and growth rate of the hake upon its capacity to biomagnify mercury?; Finally, what are the main factors governing the local variations of the mercury biomagnification in the hake subpopulations studied? The first question will be discussed using Hg_T and MeHg distributions in surface sediment and in the water column. Questions two and three will be discussed based on the hake growth parameters (total length and age), and its food web structure and trophic position, using the stable isotope ratios of carbon ($\delta\ ^{13}C$) and nitrogen ($\delta\ ^{15}N$). Answering the last question will combine the utilization of proxies for trophic position and Hg bioavailability.

■ THE EUROPEAN HAKE

The European hake (*Merluccius merluccius* Linnaeus, 1758) is broadly distributed from the coast to the continental slope of both GoL and BoB. It is the most important commercial fish species in the demersal fishery landings in the Mediterranean and the North Atlantic.[28] In the GoL its habitat extends from 30 m on the continental shelf down to 800 m on the shelf edge,[29] and it varies with age and maturity.[30−34] Young individuals are spread all along the continental shelf, while older ones live along the shelf break and at the head of the canyons. Small juvenile hake settle between 120 and 200 m depth on the continental shelf break. As they grow, hake undertake foraging migration in shallower waters and spread on the whole shelf from 30 to 200 m depth for at least two years. Adults progressively move in deeper waters and larger individuals are found in canyons on the shelf slope (200−800 m).[31,32]

The feeding habits of the hake have been described for both the Atlantic and the Mediterranean coasts.[35−38] Juveniles eat mainly suprabenthic crustaceans and small benthic fish. In the GoL it becomes more piscivorous when it reaches 15−20 cm,[38] with prey-size varying positively with the size of the hake. In the GoL, adult hake diet is mainly composed of sardine and blue whiting (>50%).[37,39] The most important prey of hake in the

BoB is the horse mackerel (45%), then juvenile hake (19%), and blue whiting (<15%).[40] In Atlantic waters hake cannibalism is common and juvenile hake may represent up to 80% of the diet of larger hake.[40]

The growth rates of the different populations were subject to intense debate until direct measurements were made, based on conventional tagging.[41] Growth parameter estimations appear to be slightly lower in the GoL than in the BoB, with females growing faster than males in both environments.[42]

■ MATERIAL AND METHODS

Sampling. Hakes and prey samples were collected in five sectors of the GoL and within the Great mud bank of the BoB (Figure 1). They were caught during bottom-trawl and gill nets between 50 and 450 m depth. A total of 440 hakes with a total length (L) of from 7 to 70 cm and 59 prey pools were sampled for Hg_T and MeHg analyses. Hakes were pretreated in the laboratory for total body weight (WW), total length (TL), and macroscopic sex determination. Seventy two (72) hakes from BoB were also collected by similar fish nets. The main characteristics of GoL and BoB are given as Supporting Information (SI 1). Water and sediment samples in GoL and BoB were also collected for chemical analysis; their locations are given in the caption of Figure 1. Details for sampling, pretreatment, and analytical procedures are given as Supporting Information (SI 2).

Chemical Analyses. Hg_T determination in biota was carried out on an aliquot section of the dried muscle by atomic absorption spectrophotometry using an automatic mercury analyzer (AMA-254, Altec) after dry digestion.[43] The accuracy and the reproducibility of the method were established using certified fish muscle reference material (DORM-1, National Research Council of Canada). The certified values (0.80 ± 0.07 mg kg^{-1}, dry weight) were reproduced (measured: 0.85 ± 0.01 mg kg^{-1}, d.w.) within the confidence limits. Repeatability varied from 2 to 7% depending on the concentration of the sample. The detection limit was 0.007 mg kg^{-1} (d.w.). Monomethylmercury (MeHg) in biota was determined after propylation by isotopic dilution with GC-ICP-MS (Thermo, X Series Quadrupole ICP-MS). Detailed procedure is given in Supporting Information (SI 2). The certified values of the CRM IAEA-436 (3.67 ± 0.42 mg kg^{-1}, d.w.) were reproduced (measured: 3.63 ± 0.23 mg kg^{-1}) within the confidence limits. Repeatability varied ca. 10%, depending on the concentration of the sample. The detection limit was 0.002 mg kg^{-1} (d.w.).

The trophic structure of the hake food web was established using stable carbon ($\delta^{13}C$) and nitrogen ($\delta^{15}N$) isotope ratios.[45] In hake and other fish species, analyses were made on dorsal white muscle. For other biota, measures were performed on the whole organism. Detailed procedure is given in Supporting Information (SI 2).

Bioenergetic Modeling. The bioaccumulation model for hake described by Bodiguel et al.[46] was used to check the potential effect of hake growth on Hg bioaccumulation in the fish. An assimilation efficiency standard for Hg of 0.8 was chosen following Trudel and Rasmussen,[47] and we considered that a hake preys on sizes smaller than half of its length.[40] The elimination rate (E) of MeHg was calculated using the following equation:[47] $\mathrm{Ln}E = 0.066T - (0.2\mathrm{Ln}W) - 5.83$ where E is the elimination rate of MeHg (l g d^{-1}), T is the water temperature (°C), and W is the weight of the fish (g). A more detailed description of the DEB model and the coupling with bioaccumulation model, based on reference 46, is given in Supporting Information (SI 3). The age of each individual was calculated using the equation age vs TL.[42] Probability calculations for R significance were performed using Vassar-Stats.[48]

■ RESULTS

Methylated Mercury in the Water and Sediment. In the water column at the shelf edge, the vertical distributions of MeHg exhibited increasing concentrations with depth, peaking in the pinocline regions, between 150 and 400 m and around 800 m for GoL and BoB, respectively (Figure 2). Furthermore,

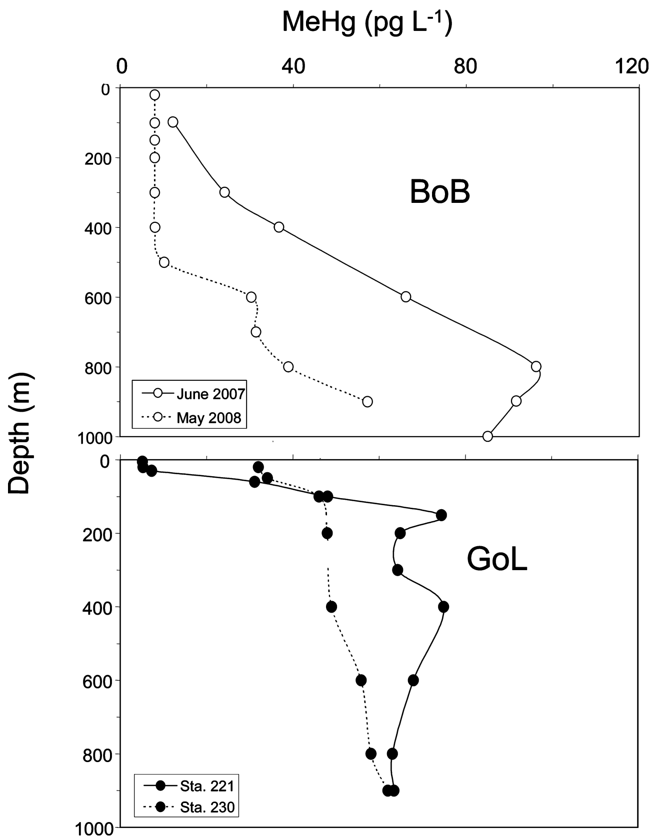


Figure 2. Methylated mercury (MeHg) in the water column of the shelf edge of Gulf of Lions (GoL) and Bay of Biscay (BoB). GoL stations 221 and 230 were located on the slope at longitudes of 4° 32.29′ E and 4° 27.96′ E, and latitudes of 42° 41.88′ N and 42° 34.71′ N, respectively. The BoB station was located at the edge of European shelf near La Chapelle bank (7° 15.40′ W; 47° 25.00′ N).

mean MeHg concentrations in the intermediate waters were lower than those in the surface waters (Table 1). In surface waters the MeHg concentrations were lower in the GoL shelf than in the open Mediterranean (Table 1). Hg_T concentrations in surface shelf sediments varied from 0.02 to 0.36 μg g^{-1} and from 0.05 to 0.40 μg g^{-1} (d.w.) for GoL and BoB, respectively. In the GoL sediment, MeHg represents less than 1% of the Hg_T (Table 1). No MeHg determination is available for BoB shelf sediments. More details on Hg distributions in sediments are given in Supporting Information (SI 4).

Allometry and the Trophic Position of Hake. The total length (TL) of hake captured ranged from 5.5 to 81 cm and from 12 to 51 cm for GoL and BoB, respectively. The WW (g) versus TL (cm) relationships were $WW = 0.0039TL^{3.16}$ (R^2 =

Table 1. Summary Statistics of Total (HgT) and Methylmercury (MeHg) in Water (Dissolved), Sediment, and Biota (Dry Weight) of the Gulf of Lions (GoL) and Bay of Biscay (BoB)[a]

	location	HgT	MeHg	MeHg/Hg_T (%)
surface waters (0−50 m) (pg L^{-1})	GoL (shelf)	292 ± 163 (87)	4.5[b] ± 2.6 (87)	1.5
	GoL (open sea)[c]	203 ± 48 (35)	6.9 ± 0.7 (33)	3.4
	BoB (shelf edge)	304 ± 97 (2)	<10 (2)	<3.2
intermediate waters (50−400 m) (pg L^{-1})	GoL (shelf edge)	336 ± 59 (8)	54 ± 16 (9)	16
	GoL (open sea)[c]	287 ± 238 (9)	33 ± 43 (9)	12
	BoB (shelf edge)	298 ± 127 (10)	12[b] ± 48 (10)	4
surface sediments (μg g^{-1})	GoL (shelf and canyons)	0.064 ± 0.037 (31)	0.00032 ± 0.00010 (31)	0.6
	BoB (shelf)[d]	range: 0.05−0.40	---	---
zooplankton (μg g^{-1})	GoL	0.099 ± 0.023 (58)	0.0123 ± 0.014 (33)	12
	BoB	0.012 ± 0.002 (2)	0.0039 ± 0.0013 (2)	33
demersal shrimp (μg g^{-1})	GoL	0.26 ± 0.12 (4)	0.17 ± 0.09 (3)	65
	BoB	0.090 ± 0.020 (25)	0.060 ± 0.009 (25)	67
dermersal fish (μg g^{-1})	GoL	0.875 ± 0.780 (12)	0.627 ± 0.512 (7)	72
	BoB	0.130 ± 0.051 (6)	0.120 ± 0.047 (6)	92
pelagic fish (μg g^{-1})	GoL	0.163 ± 0.085 (34)	0.124 ± 0.071 (32)	76
	BoB	0.084 ± 0.066 (13)	0.056 ± 0.044 (13)	67

[a]Mean ± standard deviation, number of determinations in brackets. Detailed data are given in Supporting Information, Table S1. To compare fishes of similar size the Hg concentrations have been modeled using allometric equations given in Supporting Information, SI 6. [b]Value calculated with concentrations lower than the detection limit (DL) taken as equal to half of the DL. [c]Open Mediterranean Sea data are from the same water layer from Tyrrhenian, Ionian, and Algero-Provencal basins (Refs 24 and 27). [d]Data are from OSPAR convention monitoring program (http://www.ospar.org/) and RNO network (http://envlit.ifremer.fr/surveillance). Hake's dietary preferences are given in Supporting Information (Table S3).

0.99) and WW = $0.0042TL^{3.13}$ (R^2 = 0.93) for GoL and BoB, respectively. The sex ratios (male/total) of mature captured animals were 31% and 49% for GoL and BoB, respectively. For GoL hake δ^{15}N values varied from 7.5‰ for the smallest hake to 11.9‰ for the largest ones, whereas it ranged from 11‰ to 15‰ in the smallest and the largest individuals in BoB. For all sizes, the δ^{15}N values observed were higher for hakes captured in BoB than in GoL (Supporting Information, Figure S1). Figure S2 illustrates the δ^{15}N and δ^{13}C distributions in both the GoL and the BoB food web components for the European hake. An increase in δ^{15}N and δ^{13}C from seston to hakes was observed in both environments.

Total and Methylmercury Distribution in Hake Muscle. Hg_T concentrations in hake muscle against total length are shown in Figure 3a, and summarized statistics are given in Table 2. The most striking feature was the Hg_T increase in muscle tissue with increasing fish size, a common observation in predatory fish for almost forty years.[1,49] Concentrations of Hg_T from the smallest to the largest hake followed an exponential function in GoL ($Hg_T = 0.067e^{0.69TL}$, R^2 = 0.98, $p < 0.001$), as well as in BoB ($Hg_T = 0.169e^{0.025TL}$, R^2 = 0.30, $p < 0.14$).[50] A closer observation of the GoL data set revealed a change of slope in the relationship for individuals larger than 40 cm. Such a change was lacking for BoB hake, most likely because of the lack of large-sized fishes (Table 2). Normalized for common TL and age interval Hg_T mean concentrations are significantly higher ($p < 0.001$) in GoL than in BoB hakes (Table 3). MeHg represented 65−99% and 61−91% of Hg_T of in fish muscle tissue of GoL and BoB, respectively, consistent with previous results for the same species from other parts of the Mediterranean sea (Table 2). A significant difference was observed between males and females from the GoL, with slightly higher concentrations in males (Figure 3c).

Total and MeHg Distribution in Hake Food Web. Summary statistics are given for HgT and MeHg in the biota from both GoL and BoB (Table 1). The most striking results are (i) that the proportion of Hg as MeHg increased from zooplankton to fish, and (ii) that both MeHg and HgT concentrations were higher in biota from the GoL compared to corresponding groups collected in BoB. The Hg_T concentrations at the first two levels of the food chain in the GoL varied by 1 order of magnitude from 0.014 to 0.10 mg kg^{-1} (d.w.), with MeHg representing 1% of Hg_T in seston and up to 25% in zooplankton (Figure 4 and Supporting Information, Table S1). This range is similar to what has been observed in other geographical regions (Supporting Information, Table S2). For higher trophic level prey MeHg varied from 0.04 to 1.29 mg kg^{-1} (d.w.) and MeHg/Hg_T ratios varied from 50 to 98%, for pelagic and demersal fish. Within the same genus the higher Hg_T and MeHg were associated with larger individuals. Considering the hake and its food chain as a whole, the logMeHg vs δ^{15}N relationships in GoL (slope = 0.38, R^2 = 0.89, $p < 0.001$) and BoB were both significant (slope = 0.29, R^2 = 0.91, $p < 0.001$) (Figure 4). Considering only the food chain, the relationships between logMeHg and δ^{15}N were still statistically significant (GoL: slope = 0.35, R^2 = 0.86, $p < 0.001$; BoB: slope = 0.22; R^2 = 0.83, $p < 0.04$) (Figure 4).

DISCUSSION

Bioavailability of Mercury in the GoL and BoB Waters. Because MeHg is the Hg species efficiently retained in aquatic organisms, and its biological formation results from the activity of microorganisms,[51] MeHg concentration in the surrounding environment can be considered to be a good proxy for Hg bioavailability for aquatic trophic webs. The MeHg maximum found in the shelf water column of both GoL and BOB is located at the depth of the thermohalocline, where density gradient is maximum and allows particle accumulation (including organic matter flocs and associated bacteria); the MeHg minimum is located at surface waters (including shelf surface waters) (Table 1, Figure 2). This distribution pattern suggests a net mercury methylation at the thermohalocline at the edge of the continental shelf and a demethylation at the

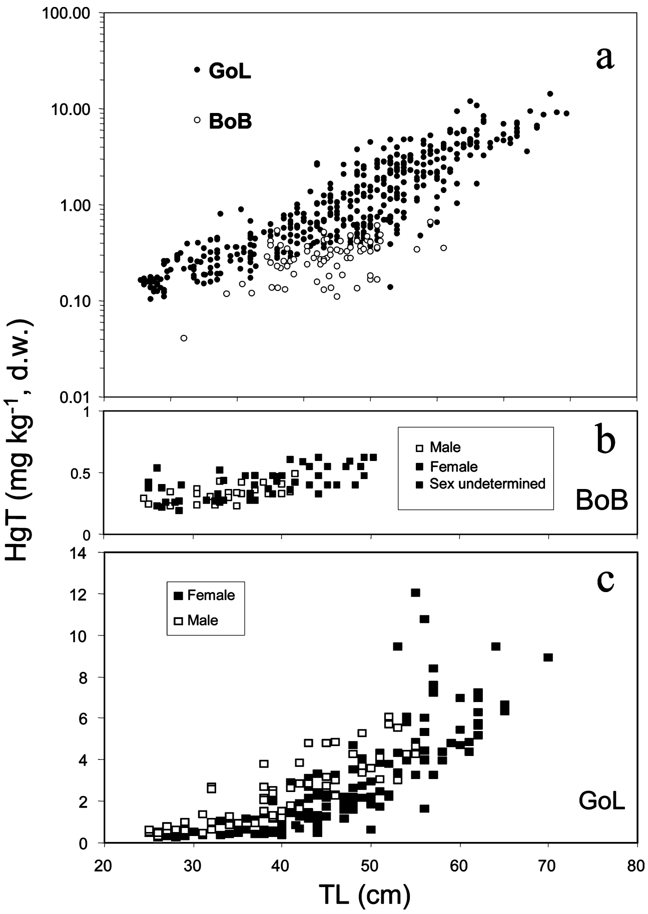


Figure 3. Relationships between total mercury concentration in muscle (Hg_T) and total length (TL) of hakes from Gulf of Lions (GoL) and Bay of Biscay (BoB). (a) Sex undetermined; (b) and (c) for male and female. With Hg_T (mg kg^{-1}) = 24.2 × 10^{-6} * TL(cm)$^{3.08}$ (R^2 = 0.80, p < 0.001) for male and Hg_T (mg kg^{-1}) = 1.41 × 10^{-6} * TL(cm)$^{3.70}$ for female (R^2 = 0.78, p < 0.001) from GoL; nonsignificant relationships for BoB hakes.

surface. This interpretation is entirely consistent with the oceanic MeHg cycling model proposed for the open Mediterranean waters[27,52] and other parts of the world Ocean,[53−55] which comprises microbiological mercury methylation in the organic matter regeneration zone and photodemethylation at surface. Thus, the exposition of marine organisms to bioavailable mercury in GoL and BoB should be minimal in surface waters and on the shelf, while maximal in deeper waters at the shelf edge. A striking difference between vertical MeHg distributions in the waters (Figure 2) is the steeper MeHg gradient from surface to depth in the GoL when compared to the BoB. This is due to the difference in water column stratification, since the thermohalocline in the GoL is nearer to the surface, due to the presence around 300 m of the high salinity Levantine Intermediate Water,[56] whereas BoB is effected by a deeper thermocline, around 600 m.[57] Thus, the organic matter regeneration zone, where MeHg is formed, is closer to the surface in the GoL than in the BoB. In their foraging areas hakes from the GoL would consequently be more exposed to MeHg than hakes from the BoB. Authors[3,58] had already noted that the mercury level in the tissues of predatory pelagic fish and their prey increased with the median depth of occurrence in the water column.

Table 3. Total Mercury (Hg_T) Concentrations in Muscle Tissue of *M. merluccius* (Mean ± Standard Deviation, Range, and Number of Determinations) for Total Length and Age Normalized Individuals[a]

	GoL	BoB
TL normalized (25−35 cm)	0.70 ± 0.51	0.28 ± 0.11
	0.28−2.71 (44)	0.11−0.54 (37)
age normalized (1−2 year)	0.52 ± 0.24	0.30 ± 0.11
	0.27−1.36 (52)	0.11−0.61 (63)

[a]The ages were calculated based on the age vs TL (ref 42).

In addition to the water column, MeHg in a coastal marine environment may also originate in the methylation of inorganic mercury in mildly reduced sediments, via microbial sulfate reduction.[59] Recently, Hollweg et al.[60] have suggested that shelf and upper slope sediments are a major source of methylmercury in the coastal ocean. Because juvenile hakes eat mainly suprabenthic crustaceans and small benthic fish,[38] the MeHg sediment source for Hg biouptake has also to be considered. Notably, suprabenthic animals are more than 5-fold MeHg depleted compared to pelagic preys (Supporting Information, Table S1). Indeed, the proportion of Hg as MeHg was ~5% within suprabenthos, while it varied between 50 and 94% with pelagic preys. Moreover, our data and data from literature tend to suggest that the shelf edge and deep sediments may be a more significant source of MeHg than continental shelf sediment (Supporting Information SI 4; refs 26 and 60). In brief, the Hg bioavailability, explored by the MeHg proxy, appears to be higher (i) in the waters of the hake habitat in GoL than in BoB, and (ii) on the shelf-edge than on the inner continental shelf of the GoL.

Effect of Trophic Position. Nitrogen stable isotope ratio (δ^{15}N) is a food-web descriptor for predatory fish that can be used as a continuous, integrative measure of trophic position, thus permitting the exploration of the length of the food chain as a governing factor for contaminant biomagnification.[61] The trophic structure of the European hake food web from GoL and

Table 2. Total Mercury (Hg_T) Concentrations in Muscle Tissue of *M. merluccius* (Mean ± Standard Deviation, Range, and Number of Determinations)[a]

	GoL (this work)	BoB (this work)	Adriatic Sea (ref 73)	Ionian Sea (ref 72)	Tyrrhenian Sea (ref 70)
Hg_T (mg kg^{-1}, d.w.)	1.67 ± 2.03	0.30 ± 0.13	0.90 ± 0.60	0.45 ± 0.40	----
	0.11−14.31 (n = 442)	0.04−0.66 (n = 71)	0.20−2.40 (n = 19)	<dl−1.50 (n = 14)	0.4−16.0 (n = 108)
MeHg/Hg_T (%)	65−100[b]	61−91[b]	60−100	73−100	----
mass range (g, w.w.)	10−2605	13−860	5−260	10−475	---
total length range (cm)	6−70	12−51	---	---	11−62

[a]Values are expressed in dry wet (d.w.) basis (the relationship $[Hg_T]_{d.w.}$ = 5 × $[Hg_T]_{w.w.}$ was used for possible conversion). [b]MeHg measurements have been performed on selected 39 and 47 sub-samples from the Gulf of Lions (GoL) and the Bay of Biscay (BoB), respectively.

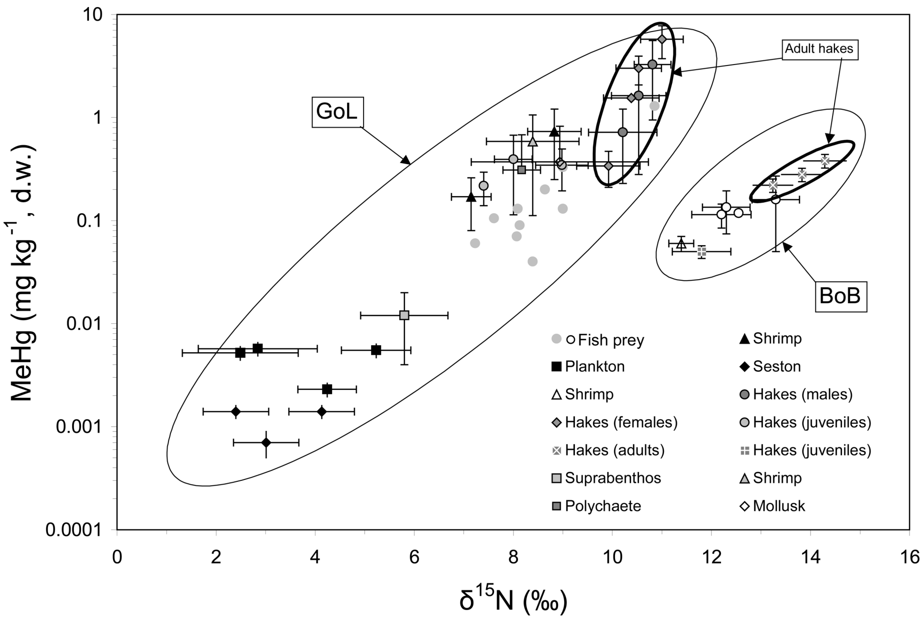


Figure 4. Relationships between δ^{15}N and monomethylmercury (MeHg) in hake muscles, preys, and food chains from Gulf of Lions (GoL) and Bay of Biscay (BoB). Means calculated are defined by zoological group for preys and as a function of size classes for hakes. GoL: $\text{Log}_{10}\text{MeHg} = 0.384\delta^{15}\text{N} - 3.925$ ($R^2 = 0.89$, $p < 0.001$); BoB: $\text{Log}_{10}\text{MeHg} = 0.287\delta^{15}\text{N} - 4.519$ ($R^2 = 0.91$, $p < 0.001$).

BoB has been explored through stable carbon and nitrogen isotope analyses in both the BoB and the GoL (Supporting Information, Figure S2). The shift in δ^{15}N does not indicate higher trophic positions for hakes from the BoB than for those from the GoL, but differences in isotopic chemistry and/or metabolism and feeding sources (Supporting Information, SI 4). Consequently, in the rest of the discussion we will not take into account δ^{15}N absolute values as a proxy for the trophic level, but discuss the result only in terms of the slope of the logMeHg vs δ^{15}N relationship, the so-called biomagnification power (BP).[62−64] A BP of 0.35 calculated for the GoL hake food web (Figure 4) is relatively high compared to other marine environments, where they vary usually between 0.16 and 0.28,[63,54] and suggests a very efficient MeHg transfer in the GoL ecosystems, which could indicate relatively low growth rates for various elements of the GoL food chains.

The BPs for hakes are 0.33 and 0.35 for GoL and BoB, respectively (Table 4). However, if we consider only hake adults, then BP is much higher for hake from the GoL (1.12) compared to those from the BoB (0.22) (Table 4, Figure 4). This difference may partially be a bias due to the lack of large

Table 4. Bioamplification Power (BP = Slope of the Relationship between δ^{15}N and Logarithm of Hg or MeHg) in Muscle of Hakes from GoL and BoB, and Other Fish[a]

		BP (R^2)	reference
Merluccius merluccius (GoL)	all individuals	0.33 (0.71*)	this work
Merluccius merluccius (GoL)	adults only	1.12 (0.92)	this work
Merluccius merluccius (BoB)	all individuals	0.35 (0.97)	this work
Merluccius merluccius (BoB)	adults only	0.22 (0.98)	this work
Mullus surmuletus (GoL)	all individuals	0.23 (0.22)	19
Mullus b. barbatus (GoL)	all individuals	0.26 (0.25)	19
Mullus b. ponticus (Black Sea)	all individuals	0.02 (0.33)	19
Gadus morhua (Arctic)	all individuals	0.09 (0.22)	67

[a]All relationships are statistically significant with $p < 0.001$, except (*) where $p < 0.02$.

specimens of high trophic positions in our BoB hake set (Table 2). However, according to the growth model by Mellon-Duval et al.,[42] the largest hakes collected in this study are 6 years old for the GoL and only 3 years old for the BoB. This increase of BP for GoL hake entering adulthood occurs when a change in feeding habits occurs, when they switch from crustaceans and benthic fish to pelagic fish at 15−20 cm TL,[38] which are 5−10 times MeHg enriched compared to crustacean and suprabenthic organisms (Supporting Information, Table 1, Table S1, Figure 4). In short, ontogenetic diet and trophic status are governing factors for the MeHg biomagnification in GoL and BoB hakes, but it does not mean that other processes such as growth rate[65,66] and geographical factors[67] can not affect the equation parameters, namely the slope and the intercept of the MeHg vs the δ^{15}N model.

Effect of Dilution by Hake Growth. Von Bertalanffy models, established with recaptured male and female tagged hakes from the GoL, present growth parameters testifying to the significantly faster growth of females compared to males, with maximal lengths ($L\infty$) of 101 and 73 cm and growth rates (k) of 0.236 and 0.239 yr^{-1}, respectively.[42] In our GoL specimen set, the largest TLs reach 55 and 70 cm for the male and the female, respectively (Figure 3), corresponding to animals 5−6 years old. Combining the two sexes, a higher growth rate of BoB hake compared to those from the GoL is also established,[42,68] with k of 0.25 and 0.18 yr^{-1}, respectively. Thus, the largest hake in our BoB samples set, with a TL of 51 cm, corresponds to half the age (3 years) of the largest from the GoL. Plotting Hg_T concentrations against age permits the accessing of changes in the bioaccumulation rate during the life of the hake (Figure 5). It clearly appears that the lowest concentrations were encountered in young hakes from the BoB (Figure 5). The most striking change in Hg_T concentrations occurs in 2-year-old specimens, which corresponds to fish entering adulthood, 29−38 cm TL, in the Mediterranean,[34] and 33−44 cm TL for the Eastern Atlantic.[69] Interestingly, this change was already noted for hakes of the same size from the

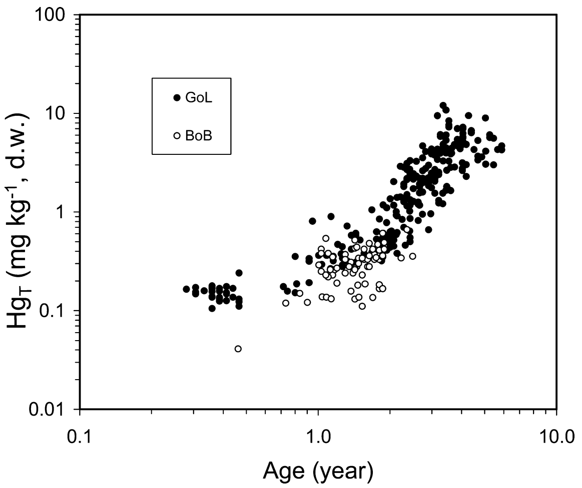


Figure 5. Relationships between total mercury concentration in muscle (Hg_T) and age for male and female hakes from Gulf of Lions (GoL) and Bay of Biscay (BoB).

Adriatic and interpreted as a result from reduction of fish growth rate.[70] In addition, the slightly higher concentrations in males compared to females from the GoL (Figure 3c), may be related to the higher growth rate of the female,[42] consistent with similar observations made for Tyhrrenian Sea hakes.[70] No such difference was noted for the BoB hakes (Figure 3b), since the lengths of most fish sampled were limited to individuals smaller than 40 cm, a range for which the growth rate difference between male and female is negligible.[41]

The bioenergetic modeling developed for contaminant bioaccumulation in hake from the BoB and the GoL[46] was applied to our Hg data set to test the effect of the growth difference between BoB and GoL hakes on the amplitude of Hg bioaccumulation in the muscle tissue of the fish, with all other modeling parameters (assimilation and elimination rate of Hg and Hg concentration in food as well) being constant. The results of the tests indicate that the difference in growth rate between the GoL and the BoB induces a muscle Hg bioaccumulation GoL/BoB ratio ranging from 1 to 2 for juveniles and the fastest growing individuals (25−35 cm), respectively (Supporting Information, Figure S2). This is enough to account for a large part of the GoL/BoB ratio (1.7−2.5) measured on size/age normalized hake mercury concentrations (Table 3).

Geographical Tendency. Table 5 gives the equations of the logHg_T vs $\delta^{15}N$ relationships in muscle tissue depending on the capture location of hake along the GoL shelf and margins. If the slope indicates the BP (see above), the interception of the equation, or Hg concentrations normalized for a common $\delta^{15}N$ value, may be used as a proxy for exploring the geographical tendency of the contamination level or Hg impregnation of the environment.[67] Generally, individuals live at least a few months in the different habitats occupied during their ontogenetic migration. This time is sufficiently long for them to incorporate the isotopic signal of their food in each habitat and region[34,35,38] and most likely also really reflect their exposure to contaminants from these regions. From the results in Table 5, it appears that the lowest Hg_T normalized concentrations (for $\delta^{15}N$ = 10 ‰) occur for the hake captured near the region of the pro-delta of the Rhône River (sectors 1 and 2) (Figure 1b). At the shelf edge (sector 5), Hg_T normalized concentrations were maximum. In other sectors, Hg_T normalized concentrations were in between or insignificant (Table 5). If the Rhône plume brings substantial inorganic Hg, mainly associated with particles,[24] to the GoL shelf, our results suggest that the bioavailable Hg for hake (i.e., MeHg) is more abundant and may be from a different source in offshore ecosystems (connected with open waters and sediments from the shelf edge) than in an inshore environment such as the Rhône pro-delta area. These results get close to those obtained with whales from the Arctic Ocean, where estuarine belugas had lowest Hg levels compared to those from the Beaufort open sea.[71] The hypothesis of differences in Hg bioavailability between habitats is supported by our finding of higher MeHg concentrations in offshore than in inshore water (Tables 1). Interestingly, studying the Hg bioaccumulation in fish from the Gulf of Mexico, using a stable isotope distribution of N, C and Hg[54,72] conclude that coastal and migratory foodwebs are largely disconnected and have different MeHg sources. Such a disconnection is more than likely in the Northwestern Mediterranean coastal ecosystems, with food webs based on terrestrial or marine particulate organic carbon.[45] We suggest, with other authors,[67,72] that MeHg is either advected from coastal environments and demethylated before entering the foodweb, or that MeHg was sourced and methylated in the open ocean.

Table 5. Parameters of the logHg_T vs $\delta^{15}N$ Equations in Hake Muscle Tissue Depending on the Capture Location along the GoL Shelf and Margins (BP = Bioaccumulation Power)

geographical sector (Figure 1b)	slope (BP)	intercept at $\delta^{15}N$ = 10 Hg_T (mg kg^{-1}, d.w.)	R^2 (n)	probability
1	0.17	−0.28	0.41 (54)	<0.001
2	0.19	−0.25	0.54 (36)	<0.001
3			0.03 (13)	0.29
4	0.18	−0.41	0.74 (27)	<0.001
5	0.40	+0.19	0.55 (128)	<0.001

ASSOCIATED CONTENT

Supporting Information

Additional information on "Study sites" (SI 1), "Material and Methods" (SI 2), "Bioenergetic and bioaccumulation modeling" (SI 3), "Total and methylmercury in sediments" (SI 4), "Carbon and nitrogen isotopes" (SI 5), "Total and methylmercury distribution in the BoB hake food web" (SI 6), "Isotopic ratios of nitrogen ($\delta^{15}N$), total mercury (Hg_T) and monomethylmercury (MeHg) in plankton and food web components" (Table S1), "Hg_T et MeHg in plankton" (Table S2), "Total length vs $\delta^{15}N$ relationships in hake muscle" (Figure S1), "Carbon and nitrogen stable isotope ratios of the main components of hake food web" (Figure S2), "Results of bioaccumulation modeling" (Figure S3), with associated references. This material is available free of charge via the Internet at http://pubs.acs.org.

AUTHOR INFORMATION

Corresponding Author

*Phone: +33 (0)4 94 30 49 14; E-mail: dcossa@ifremer.fr.

Notes

The authors declare no competing financial interest.

■ ACKNOWLEDGMENTS

Thanks are due to the French Agency for Research (ANR) and the French Institute for the Exploitation of the Sea (Ifremer), who through the MERLUMED (ANR-05-ECCO-003-02) and MEDICIS projects have funded this research.

■ REFERENCES

(1) Johnels, A. G.; Westermark, T.; Berg, W.; Persson, P. I.; Sjostrand, B. Pike (*Esox lucius* L.) and Some Other Aquatic Organisms in Sweden as Indicators of Mercury Contamination in the Environment. *Oikos* **1967**, *18*, 323−333.

(2) Jernelöv, A.; Lann, H. Mercury accumulation in Food chains. *Oikos* **1971**, *22*, 403−406.

(3) Magalhães, M. C.; Costa, V.; Menezes, G. M.; Pinho, M. R.; Santos, R. S.; Monteiro, L. R.; Murray, J. Intra-and inter-specific variability in total and methylmercury bioaccumulation by eight marine fish species from the Azores. *Mar. Pollut. Bull.* **2007**, *54*, 1654−1662.

(4) Mason, R. P.; Reinfelder, J. R.; Morel, F. M. M. The uptake, toxicity, and trophic transfer of mercury in a coastal diatom. *Environ. Sci. Technol.* **1996**, *30*, 1835−1845.

(5) Pickhardt, P. C.; Fisher, N. S. Accumulation of inorganic and monomethylmercury by freshwater phytoplankton in two contrasting water bodies. *Environ. Sci. Technol.* **2007**, *41*, 125−131.

(6) Luengen, A. C.; Flegal, A. R. Role of phytoplankton in mercury cycling in the San Francisco Bay estuary. *Limnol. Oceanogr.* **2009**, *54*, 23−40.

(7) Mason, R. P.; Laporte, J. M.; Andres, S. Factors controlling the bioaccumulation of mercury, methylmercury, arsenic, selenium, and cadmium by freshwater invertebrates and fish. *Arch. Environ. Contam. Toxicol.* **2000**, *38*, 283−297.

(8) Konovalov, Y. D. A review of the binding of cadmium and mercury in fish by proteins and low molecular thiols. *Hydrobiol. J.* **1994**, *30*, 42−51.

(9) Harris, H. H.; Pickering, I. J.; George, G. N. The chemical forms of mercury in fish. *Science* **2003**, *301*, 1203.

(10) Amlund, H.; Lundebye, A.-K.; Berntssen, M. H. G. Accumulation and Elimination of Methylmercury in Atlantic cod (*Gadus morhua* L.) Following Dietary Exposure. *Aquat. Toxicol.* **2007**, *83*, 323−330.

(11) Bloom, N. S. On the chemical form of mercury in edible fish and marine invertebrate tissue. *Can. J. Fish. Aquat. Sci.* **1992**, *49*, 1010−1017.

(12) Chen, C.; Amirbahman, A.; Fisher, N.; Harding, G.; Lamborg, C.; Nacci, D.; Taylor, D. Methylmercury in Marine Ecosystems: Spatial Patterns and Processes of Production, Bioaccumulation, and Biomagnification. *Ecohealth* **2008**, *5*, 399−408.

(13) Karimi, R.; Chen, C. Y.; Pickhardt, P. C.; Fisher, N. S.; Folt, C. L. Stoichiometric controls of mercury dilution by growth. *Proc. Natl. Acad. Sci. U.S.A.* **2007**, *104*, 7477−7482.

(14) Sharma, C. M.; Borgstrøm, R.; Sinkaberg Huitfeldt, J.; Rosseland, B. O. Selective exploitation of large pike *Esox lucius*: Effects on mercury concentrations in fish populations. *Sci. Total Environ.* **2008**, *399*, 33−40.

(15) Thibaud, Y. Teneurs en mercure de quelques poissons de consommation courante. *Sciences Pêches. Bull. Inst. Pêches Marit.* **1971**, *206*, 1−10.

(16) Bernhard, M.; Renzoni, A. Mercury concentrations in Mediterranean marine organisms and their environments: Natural and anthropogenic origins. *Thalassia Jugosl.* **1977**, *13*, 265−272.

(17) Monteiro, L. R.; Isidro, E. J.; Lopes, H. D. Mercury content in relation to sex, size, age and growth in two species of scorpionfish (*Helicolenus dactylopterus* and *Pontinus kuhlii*) from Azorean waters. *Water, Air Soil Pollut.* **1991**, *56*, 359−367.

(18) Joiris, C. R.; Holsbeek, L.; Laroussi Moatemri, N. Total and methylmercury in sardines *Sardinella aurita* and *Sardinella pilchardus* from Tunisia. *Mar. Pollut. Bull.* **1999**, *38*, 188−192.

(19) Harmelin-Vivien, M.; Cossa, D.; Crochet, S.; Bănaru, D.; Letourneur, Y.; Mellon-Duval, C. Differences in mercury bioaccumulation in red mullets from the North-western Mediterranean and Black seas. *Mar. Pollut. Bull.* **2009**, *58*, 679−685.

(20) Bernhard, M. 1985. Mercury accumulation in a pelagic food chain. In *Environmental Inorganic Chemistry*; Metertell, A. E., Irgolic, K. J., Eds.; VCH Publishers: Deerfield Beach, FL, 2000.

(21) Lahaye, L.; Bustamante, P.; Dabin, W.; van Canneyt, O.; Dhermain, F.; Cesarini, C.; Pierce, G. J.; Caurant, F. New insights from age determination on toxic element accumulation in striped and bottlenose dolphins from Atlantic and Mediterranean waters. *Mar. Pollut. Bull.* **2006**, *52*, 1219−1230.

(22) Aston, S. R.; Fowler, S. W. Mercury in the Open Mediterranean: Evidence of Contamination. *Sci. Total Environ.* **1985**, *43*, 13−26.

(23) Horvat, M.; Kotnik, J.; Logar, M.; Fajon, V.; Zvonaric, T.; Pirrone, N. Speciation of mercury in surface and deep-sea waters in the Mediterranean Sea. *Atmos. Environ.* **2003**, *37*, 93−108.

(24) Cossa, D.; Coquery, M. The Mediterranean mercury anomaly, a geochemical or a biological issue. In *The Mediterranean Sea. Handbook of Environmental Chemistry*, Vol 5; Saliot, A., Ed.; Springer, 2005; pp 177−208.

(25) Pirrone, N.; Barth, H. Mercury Cycling in Surface and Deep Waters of the Mediterranean Sea. *Mar. Chem.* **2007**, *107*, 1−116.

(26) Ogrinc, N.; Monperrus, M.; Kotnik, J.; Fajon, V.; Vidimova, K.; Amouroux, D.; Kocman, D.; Tessier, E.; Žižek, S.; Horvat, M. Distribution of mercury and methylmercury in deep-sea surficial sediments of the Mediterranean Sea. *Mar. Chem.* **2007**, *107*, 31−48.

(27) Cossa, D.; Averty, B.; Pirrone, N. The origin of methylmercury in the open Mediterranean water column. *Limnol. Oceanogr.* **2009**, *54*, 837−844.

(28) Aldebert, Y.; Recascens, L.; Lleonart, J. Analysis of Gear Interactions in a Hake Fishery: the Case of the Gulf of Lions (NW Mediterranean). *Sci. Mar.* **1993**, *57*, 207−217.

(29) Farrugio, H.; Marin, C. *Etat des pêcheries et des stocks de poissons démersaux et de petits pélagiques du golfe du Lion (Unités de gestion CGPM 37 1.2)*; Travaux du laboratoire RH de l'IFREMER pour la période 1983−1999; Document de travail CSC/CGPM, Rome, 7−10 juin 1999.

(30) Recasens, L.; Lombarte, A.; Morales-Nin, B.; Torres, G. J. Spatiotemporal variation in the population structure of the European hake in the NW Mediterranean. *J. Fish Biol.* **1998**, *53*, 387−401.

(31) Orsi-Relini, L.; Papaconstantinou, C.; Jukic-Peladic, S.; Souplet, A.; Gil de Sola, L.; Piccinetti, C.; Kavadas, S.; Rossi, M. Distribution of the Mediterranean hake populations (*Merluccius merluccius smiridus* Rafinesque, 1810) (Osteichthyes: Gadiformes) based on six years monitoring by trawl surveys: Some implications for management. *Sci. Mar.* **2002**, *66* (Suppl. 2), 21−38.

(32) Maynou, F.; Lleonart, J.; Cartes, J. E. Seasonal and spatial variability of hake (*Merluccius merluccius* L.) recruitment in the NW Mediterranean. *Fish. Res.* **2003**, *60*, 65−78.

(33) Maynou, F.; Olivar, M. P.; Emelianov, M. Patchiness of eggs, larvae and juveniles of European hake *Merluccius merluccius* from the NW Mediterranean. *Fish. Oceanogr.* **2006**, *15*, 390−401.

(34) Goñi, R.; Adlerstein, S.; Alvarez, F.; Garcia, M.; Sanchez, P.; Sbrana, M.; Maynoud, F.; Viva, C. Recruitment indices of European hake, *Merluccius merluccius* (Linnaeus 1758), in the Northwest Mediterranean based on landings from bottom-trawl multispecies fisheries. *ICES J. Mar. Sci.* **2004**, *61*, 760−773.

(35) Guichet, R. The diet of European hake (*Merluccius merluccius*) in the northen part of the bay of Biscay. *ICES J. Mar. Sci.* **1995**, *52*, 21−31.

(36) Papaconstantinou, C.; Caragitsou, E. The food of hake *Merluccius merluccius* in the Greek sea. *Vie Milieu* **1987**, *37*, 77−83.

(37) Bozzano, A.; Recasens, L.; Sartor, P. Diet of the European hake *Merluccius merluccius* (Pisces: Merlucciidae) in the Western Mediterranean (Gulf of Lion). *Sci. Mar.* **1997**, *61*, 1−8.

(38) Ferraton, F.; Harmelin-Vivien, M.; Mellon-Duval, C.; Souplet, A. Does spatio-temporal variation in diet affect the condition and abundance of European hake (*Merluccius merluccius*) juveniles in the Gulf of Lions (NW Mediterranean)? *Mar. Ecol.: Prog. Ser.* **2007**, *337*, 197−208.

(39) Mellon-Duval, C. Unpublished data, Ifremer, Centre de Recherche Halieutique Méditerranéenne et Tropicale, BP 171, F-34203 Sète, France.

(40) Mahe, K.; Amara, R.; Bryckaert, T.; Kacher, M.; Brylinski, J. M. Ontogenetic and spatial variation in the diet of hake (*Merluccius merluccius*) in the bay of Biscay and the Celtic Sea. *ICES J. Mar. Sci.* **2007**, *64*, 1210−1219.

(41) de Pontual, H.; Groison, A. L.; Pineiro, C.; Bertignac, M. 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.* **2006**, *63*, 1674−1681.

(42) Mellon-Duval, C.; de Pontual, H.; Métral, L.; Quemener, L. Growth of European hake (*Merluccius merluccius*) in the Gulf of Lions based on conventional tagging. *ICES J. Mar. Sci.* **2009**, *67*, 62−70.

(43) Cossa, D.; Coquery, M.; Nakhlé, K.; Claisse, D. *Dosage du mercure total et du monométhylmercure dans les organismes et les sédiments marins*; Méthodes d'analyse en milieu marin. Publication Ifremer, Brest; ISBN 2-84433-105-X, 2002.

(44) Rodriguez Martin-Doimeadios, R. C.; Monperrus, M.; Krupp, E.; Amouroux, D.; Donard, O. F. X. Using speciated isotope dilution with GC-inductively coupled plasma MS to determine and unravel the artificial formation of monomethylmercury in certified reference sediments. *Anal. Chem.* **2003**, *75*, 3202−3211.

(45) Darnaude, A.; Salen-Picard, C.; Polunin, N. V. C.; Harmelin-Vivien, M. Trophodynamic linkage between river runoff and coastal fishery yield elucidated by stable isotope data in the Gulf of Lions (NW Mediterranean). *Oecologia* **2004**, *138*, 325−332.

(46) Bodiguel, X.; Maury, O.; Mellon-Duval, C.; Rouspsard, F.; Le Guellec, A. M.; Loizeau, V. A dynamic and mechanistic model of PCB bioaccumulation in the European hake (*Merluccius merluccius*). *J. Sea Res.* **2009**, *62*, 124−134.

(47) Trudel, M.; Rasmussen, J. B. Predicting mercury concentration in fish using mass balance models. *Ecol. Appl.* **2001**, *11*, 517−529.

(48) VassarStats 2001. (file:///F:/Modeles%20Logiciels%20Aide-calculs/VassarStats%20Statistical%20Computation%20Web%20Site.htm).

(49) Cutshall, N. H.; Naidu, J. R.; Pearcy, W. G. Mercury concentrations in Pacific Hake, *Merluccius productus* (Ayres), as a Function of Length and Latitude. *Science* **1978**, *200*, 1489−1491.

(50) Cossa, D.; Auger, D.; Averty, B.; Luçon, M.; Masselin, P.; Sanjuan, J. *Niveaux de concentration en métaux, métalloïdes et composés organochlorés des produits de la pêche côtière française*; Publication Ifremer, Brest; ISBN 2905434-29-5, 1991.

(51) Baldi, F. Microbial Transformation of Mercury Species and their Importance in the Biogeochemical Cycle of Mercury. In *Metal Ions in Biological systems*, Vol. *34*: *Mercury and Its Effects on Environment and Biology*; Sigel, A., Sigel, H., Eds.; Marcel Dekker: New York, 1997; pp 213−248.

(52) Heimbürger, L.-E.; Cossa, D.; Marty, J.-C.; Migon, C.; Averty, B.; Dufour, A.; Ras, J. Methylmercury distributions in relation to the presence of nano and picophytoplankton in an oceanic water column (Ligurian Sea, North-western Mediterranean). *Geochim. Cosmochim. Acta* **2010**, *74*, 5549−5559.

(53) Sunderland, E. M.; Krabbenhoft, D. P.; Moreau, J. W.; Strode, S. A.; Landing, W. M. Mercury sources, distribution, and bioavailability in the North Pacific Ocean: Insights from data and models. *Global Biogeochem. Cycles* **2009**, *23*, GB2010 DOI: 10.1029/2008GB003425.

(54) Senn, D. B.; Chesney, E. J.; Blum, J. D.; Bank, M. S.; Maage, A.; Shine, J. P. Stable isotope (N, C, Hg) study of methylmercury sources and trophic transfer in the northern Gulf of Mexico. *Environ. Sci. Technol.* **2010**, *44*, 1630−1637.

(55) Cossa, D.; Heimbürger, L.-E.; Lannuzel, D.; Rintoul, S. R.; Butler, E. C. V.; Bowie, A. R.; Averty, B.; Watson, R. J.; Remenyi, T. Mercury in the Southern Ocean. *Geochim. Cosmochim. Acta* **2011**, *75*, 4037−4052.

(56) Durrieu de Madron, X.; Nyffeler, F.; Godet, C. H. Hydrographic structure and nepheloid spatial distribution in the Gulf of Lions continental margin. *Cont. Shelf Res.* **1990**, *10*, 915−929.

(57) Fraile-Nuez, E.; Plaza, F.; Hernandez-Guerra, A.; Vargas-Yanez, M; Lavı, A. Mass transport in the Bay of Biscay from an inverse box model. *J. Geophys. Res.* **2008**, *113*, C06023 DOI: 10.1029/2007JC004490.

(58) Choy, C. A.; Popp, B. N.; Kaneko, J. J.; Drazen, J. C. The influence of depth on mercury levels in pelagic fishes and their prey. *Proc. Natl. Acad. Sci. U.S.A.* **2009**, *106*, 13865−13869.

(59) Fitzgerald, W. F.; Lamborg, C. H.; Hammerschmidt, C. R. Marine biogeochemical cycling of mercury. *Chem. Rev.* **2007**, *107*, 641−662, DOI: 10.1021/cr050353m.

(60) Hollweg, T. A.; Gilmour, C. C.; Mason, R. P. Mercury and methylmercury cycling in sediments of the mid-Atlantic continental shelf and slope. *Limnol. Oceanogr.* **2010**, *55*, 2703−2722.

(61) Cabana, G.; Rasmussen, J. B. Modeling food-chain structure and contaminant bioaccumulation using stable nitrogen isotopes. *Nature* **1994**, *372*, 255−257.

(62) Jarman, W. M.; Hobson, K. A.; Sydeman, W. J.; Bacon, C. E.; McLaren, E. B. Influence of Trophic Position and Feeding Location on Contaminant Levels in the Gulf of the Farallones Food Web revealed by Stable Isotope Analysis. *Environ. Sci. Tchnol.* **1996**, *30*, 654−660.

(63) Atwell, L.; Hobson, K. A.; Welch, H. E. Biomagnification and bioaccumulation of mercury in an Arctic marine food web: insight from stable nitrogen isotope analysis. *Can. J. Fish. Aquat. Sci.* **1998**, *55*, 1114−1121.

(64) Al-Reasi, H. A.; Ababneh, F. A.; Lean, D. R. Evaluating Mercury Biomagnification in Fish from a Tropical Marine Environment, Using Stable Isotopes (δ^{13}C and δ^{15}N). *Environ. Toxicol. Chem.* **2007**, *26*, 1572−1581.

(65) Stafford, C. P.; Haines, T. A. Mercury contamination and growth rate in two piscivore populations. *Environ. Toxicol. Chem.* **2001**, *20*, 2099−2101.

(66) Simoneau, M.; Lucotte, M.; Garceau, S.; Laliberté, D. Fish growth rates modulate mercury concentrations in walleye (*Sander vitreus*) from eastern Canadian lakes. *Environ. Res.* **2005**, *98*, 73−82.

(67) Stern, G. A.; Macdonald, R. W. Biogeographic Provinces of Total and Methymercury in Zooplankton and Fish from the Beaufort and Chukchi Seas: Results from the SHEBA Drift. *Environ. Sci. Technol.* **2005**, *39*, 4707−4717.

(68) Aldebert, Y.; Recasens, L. Estimation de la croissance du merlu du Golfe du Lion par l'analyse des fréquences de tailles. *Rapp. Comm. Int. Mer Méditerr.* **1995**, *34*, 236−240.

(69) Piñeiro, C.; Sainza, M. Age estimation, growth and maturity of the European hake (*Merluccius merluccius* (Linnaeus, 1758)) from Iberian Atlantic waters. *ICES J. Mar. Sci.* **2003**, *60*, 1086−1102.

(70) Barghigiani, C.; Ristori, T.; Biagi, F.; de Ranieri, S. Size related mercury accumulation in edible marine species from an area of the Northern Tyrrhenian Sea. *Water, Air Soil Pollut.* **2000**, *124*, 169−176.

(71) Loseto, L. L.; Stern, G. A.; Deibel, D.; Connelly, T.; Lean, D. R. S.; Prokopowicz, A.; Fortier, L.; Ferguson, S. H. Linking mercury exposure to habitat and feeding behaviour of beluga whales. *J. Mar. Syst.* **2008**, *74*, 1012−1024.

(72) Senn, D. B.; Chesney, E. J.; Blum, J. D.; Shine, J. P. Mercury bioaccumulation in coastal and migratory foodwebs in the Gulf of Mexico. Goldschmidt Conference Davos, Switzerland. Abstract in *Geochim. Cosmochim. Acta* **2009**, *73*, 13S, SA1197.

(73) Storelli, M. M.; Storelli, A.; Giacominelli-Stuffler, R.; Marcotrigiano, G. O. Mercury speciation in the muscle of two commercially important fish, hake (*Merluccius merluccius*) and striped mullet (*Mullus barbatus*) from the Mediterranean Sea: Estimated weekly intake. *Food Chem.* **2005**, *89*, 295−300.