



Organochlorine pesticides, polychlorinated biphenyls and trace elements in wild European sea bass (*Dicentrarchus labrax*) off European estuaries

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ABSTRACT

Polychlorinated biphenyls (PCBs) and organochlorine pesticides like dichloro-diphenyl-trichloroethane (DDTs), hexachlorocyclohexanes (HCHs), aldrin, dieldrin and trace elements (Cd, Cu, Se, Pb, Zn and Hg) were analysed in the muscle of European sea bass (*Dicentrarchus labrax*) sampled in Atlantic coastal regions near several important European river mouths (Gironde, Charente, Loire, Seine and Scheldt). High contamination levels were measured in the muscles of European sea bass sampled in the coastal regions near those river mouths (e.g. Σ ICES PCB = 133–10,478 $\mu\text{g kg}^{-1}$ lw and Hg = 250–2000 $\mu\text{g kg}^{-1}$ dw).

The Scheldt and the Seine are still among the most contaminated estuaries in Europe. Each region presented their specific contamination patterns reflecting different sources due to the input of the respective rivers. As fish and fishery products are the main contributors of the total dietary intake of organochlorinated pollutants, regular consumption of European sea bass with the reported contamination levels may represent a significant exposure route for the general human population.

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1. Introduction

European sea bass (*Dicentrarchus labrax*) is a euryhaline species that inhabits estuaries, lagoons and coastal waters. This fish species is carnivorous, feeding on fish, crustaceans and cephalopods (Pickett and Pawson, 1994) and thus accumulates through its food major organic pollutants, such as polychlorinated biphenyls (PCBs) and organochlorine pesticides (Loizeau et al., 2001) as well as non-essential and essential elements (Dural et al., 2006; Türkmen et al., 2005). The species is considered as a bioindicator of marine pollution (Loizeau, 2001) and displays both economic and environmental importance. Indeed, human consumption of European sea bass through fishing (8528 t in 2008) and aquaculture (66,738 t in 2008) is very important (FAO, 2008). Moderate to high levels of organochlorine compounds (Antunes and Gil, 2004; Carubelli et al., 2007; Pastor et al., 1996; Schnitzler et al., 2008; Trocino et al., 2007) and trace elements (Dugo et al., 2006; Dural et al., 2006; Durrieu et al., 2005; Miramand et al., 2001) were previously described in sea bass, which were below the European norm. It is noteworthy that the norm of PCBs for meat products (200 ng g^{-1} lipid weight (lw); (Belgisch Staatsblad-Moniteur Belge, 1999)), is not applied to fish, but instead a norm of 75 ng g^{-1} wet weight (ww) is used.

European sea bass use estuaries and coastal bay as nursery area, after they reach a mean length of 36 cm, they disperse in coastal marine

environment (Pawson et al., 2007). So European sea bass below 36 cm should be a good bioindicator organism reflecting the local water environmental pollution in their tissues. Meanwhile, the Current Minimum Legal size for fishing sea bass is 36 cm and all fish below this size should be returned into sea.

For many years seafood such as fish, molluscs and crustaceans has often been the focus of attention in nutritional and toxicological work. Nutritionists consider these products to be an important source of high-quality proteins, essential elements and (omega-3) fatty acids such as eicosapentaenoic acid (EPA) and docosahexaenoic acid (DHA). Toxicologists tend to regard seafood as a major vector for toxic substances such as toxic metals and persistent organic pollutants (Domingo and Bocio, 2007; Voorspoels et al., 2008). It is thus interesting to evaluate the nutritional benefits of nutritive elements in food and the health risks related to toxic substances, as high rates of pollutants in fish might counterbalance the health benefit derived from the essential fatty acid in fish lipid. A recent study revealed that European sea bass contribute to 7.12% of the recommendation for essential fatty acids such as EPA and DHA for inhabitants of French coastal areas (Le Blanc et al., 2006).

There is a need for more information on contaminant levels in European fish. The present study aimed to assess and compare the levels of organochlorinated compounds, non-essential and essential trace elements measured in muscle of sea bass near the mouth of several European rivers (Gironde, Charente, Loire, Seine and Scheldt). The sedentary habits of juvenile European sea bass allowed us to compare the local pollution of the different regions. The conception of the study, assessing simultaneously organic pollutants, such as PCBs and

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organochlorine pesticides as well as non-essential and essential elements is genuine. Benefits and potential risk linked to human consumption of this edible fish will be discussed.

2. Methods

2.1. Sampling

Eighty-seven European sea bass were collected between September 20th and November 1st in 2007 during different scientific missions of CEMAGREF (Institut de recherche pour l'ingénierie de l'agriculture et de l'environnement, France), IFREMER (Institut Français de Recherche pour l'Exploitation de la Mer, France) and INBO (Instituut voor Natuur-en Bosonderzoek, Belgium). Fish were caught in the coastal region near these European rivers: Gironde (France), Charente (France), Loire (France), Seine (France) and Scheldt (Belgium and The Netherlands).

The caught fish were immediately dissected. Total length and the weight were measured. Gender and maturity stage were assessed through examination of the gonads. Approximately 30 g of skeletal muscle was excised from the area behind the head, dorsal to the lateral line and anterior to the dorsal fin. The muscle samples were stored frozen at -70°C until analysis.

2.2. Chemical analysis

All solvents were of pesticide grade; n-hexane and acetone (Burdick & Jackson brand) were purchased from Fluka (Buchs, Switzerland). The Mirex (dodecachloropentacyclo-[5.3.0.0.0.0]decane) used as internal standard, the pure PCB congeners (IUPAC nos. 28, 44, 52, 66, 70, 87, 95, 101, 105, 110, 118, 128, 138, 149, 153, 156, 170, 180, 183, 187, 194, 195, 206, and 209) and the congener used as surrogate (IUPAC no. 112), were obtained from Ultra Scientific® (Wesel, Germany) and Dr Ehrenstorfer® (Augsburg, Germany). All other chemicals used were of analytical grade.

Extracts of 10 g of dorsal muscular tissue of each fish were lyophilized over 20 h and dry matter was determined gravimetrically. The lipids were extracted using an accelerated solvent extractor (ASE) (Dionex ASE 2000, Dionex Corporation) and were analysed for polychlorinated biphenyls (PCBs), dichloro-diphenyl-trichloroethane (DDTs), hexachlorocyclohexanes (HCHs), aldrin and dieldrin by gas chromatography using a Thermo Quest Trace 2000 gas chromatograph equipped with a 63Ni ECD (Thermo Quest, Trace 2000). The details of sample preparation and clean-up are provided in Schnitzler et al. (2008).

The quality control was performed by analysis of procedural blanks, by injection of standard and n-hexane blanks. Standard reference material SRM 1946 (PCBs and organochlorine pesticides in Lake Superior fish tissue) and BCR RM 349 (cod liver) were used to test the whole procedure of the analytical method accuracy (recovery efficiency for certified concentrations between 71% and 128% according to the compounds). Procedural blanks and laboratory made quality control were run with each set of samples to control the extraction and clean-up procedures. Milk cream enriched with a defined concentration of PCBs was used as a quality control for analysis. For each PCB congener and organochlorine pesticides, recovery efficiency was calculated on the basis of the concentration of the surrogate marker PCB 112. All results were corrected to obtain 100% recovery. However, the results of the PCBs and organochlorine pesticides analyses were accepted only if the recoveries were between 70% and 130%. Total PCB concentrations (ΣPCBs) were calculated as the sum of all individual quantified congeners, total International Council for the Exploration of the Sea PCBs ($\Sigma\text{ICES PCB}$) as the sum of the 7 tracer PCB congeners (IUPAC 28, 52, 101, 118, 138, 153 and 180), total DDTs (ΣDDTs) as the sum of pp'-DDT, pp'-DDE and pp'-DDD concentrations and total HCHs (ΣHCHs) as the sum of α -HCH, β -HCH and γ -HCH concentrations. For each PCB congeners and organochlorine pesticide, the limits of

quantification were $0.08\text{ }\mu\text{g kg}^{-1}$. The efficiency recovery for PCB congeners and organochlorine pesticides was always above 75%.

2.3. Trace element analysis

After being weighed and dried at 60°C to a constant weight, 2 g of muscle samples were digested in Teflon tubes with concentrated nitric acid, deionised water and H_2O_2 in a microwave oven (during 20 min between 0 and 600 W). After cooling, samples were diluted to 50 ml with deionised water in a volumetric flask. Samples for Cd, Cu, Se, Pb, and Zn were analysed by Inductively Coupled Plasma-Mass Spectrometer (ICPMS Elan DRC II) immediately after digesting. Samples for Hg were analysed by Direct Mercury Analyzer (DMA Milestones). A mean water content of $74.0 \pm 4.1\%$ was calculated in our samples. Concentrations are expressed in $\mu\text{g kg}^{-1}$ dry weight (dw). Parallel to samples, a set of certified control material samples (DOLT-3 liver and DORM-2 muscle, National Research Council Canada) went through each set of analyses to ensure the accuracy and precision of the method. Recoveries for control materials ranged from 92% to 109% for Cd, Ni, Cu, Zn, Se, and Pb. Instrumental detection limits were: Cu, $0.020\text{ }\mu\text{g kg}^{-1}$; Zn, $0.042\text{ }\mu\text{g kg}^{-1}$; Se, $0.166\text{ }\mu\text{g kg}^{-1}$; Cd, $0.005\text{ }\mu\text{g kg}^{-1}$; Pb, $0.002\text{ }\mu\text{g kg}^{-1}$; Hg, $1\text{ }\mu\text{g kg}^{-1}$, respectively. All samples showed element concentrations above the detection limits.

2.4. Statistical analysis

Statistical analysis of the data was performed using SPSS for Mac® software (SPSS Inc., version 16.0.2). The Kolmogorov-Smirnov test was used to test for normality of the statistically treated variables, and to ensure the utilisation of adapted tests. Before all individuals were regrouped for the further statistical analysis, the influence of factors like sex, weight and length was tested. The non-parametric Mann-Whitney U-test was used to compare differences in organic and trace element compound concentrations among sexes and Spearman correlation test followed by Fishers Omnibus post hoc tests were used to compare differences in organic and trace element compound concentrations in relation to length and weight. Intersite comparison of the contamination patterns was realised using discriminant analysis to assess the ability of organic and trace element concentrations to discriminate among the different collection locations. Results were judged significant when $p < 0.05$.

3. Results

3.1. Sampling

Mean body length was $31 \pm 4.6\text{ cm}$ and weight $376 \pm 190\text{ g}$. There were no significant differences in mean body length and weight among study location groups. All sea bass were estimated juvenile; gross observation of the gonads confirmed that all sea bass were immature. There was an overall sex ratio of 1:1.

The lipid proportion in the muscles of the European sea bass ranged from 0.1 to 3.9% of the wet weight with a mean value of 0.7%. Yet the lipid content did not vary significantly among the sampling locations (ANOVA; $p = 0.059$). Sea bass from the coastal region near the Gironde showed the highest lipid content in their muscles (mean value of 1.7%) and the sea bass from the coastal region near the Charente had the lowest lipid content in their muscles (mean value of 0.4%) (Table 1).

No significant difference in contaminant concentrations could be detected between the sexes (Mann-Whitney; $p > 0.05$) and no significant relationship between length and weight, and contaminant concentrations could be revealed (Spearman correlation tests followed by Fisher Omnibus post-hoc test; $p > 0.05$).

Table 1
Lipid proportion, mean contamination levels of the 27 measured PCBs, the 7 ICES PCBs, the sum of DDTs (*pp'*-DDT, *pp'*-DDE, *pp'*-DDD), the sum of HCHs and the sum of aldrin and dieldrin in the white muscle of European sea bass collected from coastal regions near several important European river mouths. The concentrations are given in $\mu\text{g kg}^{-1}$ wet weight and lipid weight (mean \pm standard deviation, (median) and min–max) * significant ANOVA; ^{a,b,c} significant after Tukey's multiple comparisons.

Coastal region	Gironde	Charente	Loire	Seine	Scheldt	Probability
n	8	8	34	26	11	
<i>Wet weight</i>						
Lipid (%)	1.7 \pm 1.1 (1.4) 0.5–3.2	0.4 \pm 0.4 (0.2) 0.1–1.2	0.9 \pm 0.7 (0.7) 0.3–3.9	0.9 \pm 0.3 (0.9) 0.1–1.4	0.6 \pm 0.6 (0.4) 0.1–2.1	F(4,83) = 2.6 p = 0.059
Σ PCB ($\mu\text{g kg}^{-1}$)	49.7 \pm 23.4 ^a (55.8) 15.1–82.3	18.1 \pm 10.0 ^b (19.8) 10.5–29.5	40.4 \pm 19.1 ^{a,b} (36.5) 17.2–101.5	46.4 \pm 21.6 ^a (41.9) 15.8–101.8	42.5 \pm 31.4 ^{a,b} (34.5) 17.2–124.7	F(4,83) = 2.7 p = 0.035*
Σ ICES PCB ($\mu\text{g kg}^{-1}$)	26.8 \pm 13.1 ^a (29.9) 7.7–45.0	7.9 \pm 4.6 ^b (8.6) 3.7–13.6	14.6 \pm 8.7 ^{b,c} (11.3) 5.7–45.9	26.0 \pm 11.6 ^{a,c} (23.8) 8.4–54.7	17.4 \pm 14.2 ^{a,b,c} (11.8) 6.2–53.1	F(4,83) = 7.5 p < 0.001*
Σ <i>p'</i> -DDT ($\mu\text{g kg}^{-1}$)	2.2 \pm 0.9 ^{a,c} (2.4) 0.8–3.3	0.4 \pm 0.3 ^b (0.5) 0.2–0.8	1.8 \pm 1.5 ^{a,b,c} (1.4) 0.4 \pm 0.2 ^b	1.2 \pm 0.6 ^{b,c} (1.2) 0.2–2.3	3.0 \pm 2.2 ^a (1.9) 1.3–7.9	F(4,83) = 5.6 p = 0.001
Σ HCH ($\mu\text{g kg}^{-1}$)	0.1 \pm 0.1 ^a (0.1) 0.0–0.2	0.2 \pm 0.1 ^{a,b} (0.2) 0.0–0.3	0.4 \pm 0.2 ^b (0.4) 0.0–0.7	0.1 \pm 0.2 ^a (0.1) 0.0–0.8	0.1 \pm 0.0 ^a (0.1) 0.1–0.2	F(4,83) = 13.1 p < 0.001*
Σ aldrin–dieldrin ($\mu\text{g kg}^{-1}$)	0.3 \pm 0.2 ^a (0.4) 0.0–0.5	0.2 \pm 0.1 ^a (0.2) 0.1–0.5	0.7 \pm 0.1 ^a (0.6) 0.1–1.9	0.5 \pm 0.3 ^{a,b} (0.4) 0.1–1.2	0.3 \pm 0.5 ^a (0.1) 0.0–1.8	F(4,83) = 3.6 p = 0.010*
<i>Lipid weight</i>						
Σ PCB ($\mu\text{g kg}^{-1}$)	1821 \pm 2102 (2422) 296–8487	3756 \pm 3077 (4546) 1550–13,917	4473 \pm 2783 (4217) 1696–20,185	4716 \pm 2880 (4500) 615–10,478	6564 \pm 3102 (6188) 2776–15,383	F(4,83) = 1.1 p = 0.364
Σ ICES PCB ($\mu\text{g kg}^{-1}$)	879 \pm 1062 ^a (1228) 133–5765	1361 \pm 1129 ^{a,b} (1421) 661–4382	1374 \pm 550 ^{a,b} (1411) 579–5229	2543 \pm 1550 ^c (2550) 616–10,478	2628 \pm 1239 ^c (2557) 1407–5379	F(4,83) = 4.4 p = 0.003*
Σ <i>p'</i> -DDT ($\mu\text{g kg}^{-1}$)	91 \pm 101 ^a (97) 18–374	92 \pm 123 ^a (108) 44–561	144 \pm 176 ^b (128) 1–691	130 \pm 113 ^b (122) 28–534	395 \pm 324 ^c (392) 131–1219	F(4,83) = 14.0 p < 0.001*
Σ HCH ($\mu\text{g kg}^{-1}$)	4 \pm 3 ^a (4) 1–10	54 \pm 43 ^c (51) 14–167	36 \pm 29 ^b (40) 6–183	9 \pm 25 ^a (7) 1–197	15 \pm 12 ^{a,b} (16) 3–52	F(4,83) = 5.4 p = 0.001*
Σ aldrin–dieldrin ($\mu\text{g kg}^{-1}$)	27 \pm 16 (29) 9–71	48 \pm 28 (82) 13–122	52 \pm 68 (59) 9–438	47 \pm 49 (44) 10–276	11 \pm 20 (16) 0–85	F(4,83) = 1.9 p = 0.114

3.2. Organochlorinated compound analysis

The levels of organochlorinated compounds, (PCBs and pesticides on ww basis) varied significantly among sampling locations (ANOVA; $p < 0.05$). The general contamination trend showed higher levels in the coastal regions near the Seine and the Gironde followed by the Scheldt and Loire, while the lowest levels were observed in the coastal region near the Charente (Table 1). On a lipid weight basis, Σ of PCB concentrations did not differ between the sampling areas whereas other OCs (for Σ of International Council for the Exploration of the Sea (ICES) PCBs, Σ of DDTs, Σ of Pesticides and Σ of HCHs) differed significantly (ANOVA; $p < 0.05$). The data, expressed in ng g^{-1} lipid weight, shows different trends, here the coastal regions near the rivers Scheldt and Seine are the most contaminated, followed by the Loire and the Charente and finally the Gironde is the less contaminated (Table 1).

The 7 ICES (International Council for the Exploration of the Sea) PCBs (IUPAC 28, 52, 101, 118, 138, 153 and 180), the major congeners and most predominantly used in the different commercial mixtures of PCBs, accounted for 40–60% of all the found congeners according to the origin of the samples. They were found in European sea bass in decreasing importance: 138 > 153 > 180 > 101 > 52 > 118 > 28 with only minor changes among sampling regions.

Contamination pattern differences between sampling locations were explored by discriminant analysis to show how several predefined groups of individuals (sampling location of sea bass) may be separated by given measurements of several variables (Fig. 1A). It provides linear functions of variables that best separate the cases into the predefined groups. The five sea bass groups could be discriminated by their

contamination pattern (lipid weight basis), principally by the compounds β -HCH, aldrin, dieldrin, IUPAC PCB 70, 101, 87, 149, 153, 180, 170, 194, *pp'*-DDT, *pp'*-DDE and *pp'*-DDD. The first discriminant function (root) explained 36% of the variations between groups involving mostly β -HCH, dieldrin, IUPAC PCB 70, 101, 87, 153, 194, *pp'*-DDE and *pp'*-DDD, and the second discriminant function explained a further 31% of the variation between groups, involving the aldrin, IUPAC PCB 149, 180, 170 and *pp'*-DDT concentrations. Together, the two discriminant functions explained 67% of the variance. The plot of the discriminant analysis is shown in Fig. 1A. Each sampling region had his well defined contamination pattern; the 5 groups are well separated from each other.

3.3. Trace element analysis

All tested metals, including Hg, Pb and Cd, were found in detectable quantities in all samples (Table 2). Several trace element concentrations (Zn, Se, Hg, Pb, and Cd) varied significantly between the sampling locations (ANOVA; $p < 0.05$). No clear contamination pattern could be explored by discriminant analysis, as the sampling locations reveal no clear contamination trend. Instead all individuals were regrouped together in the centre of the discriminant analysis plot (Fig. 1B).

4. Discussion

4.1. OC contamination level

Various authors suggested normalising concentrations of pollutants to the lipid contents in order to reduce intra-species variability (Pastor

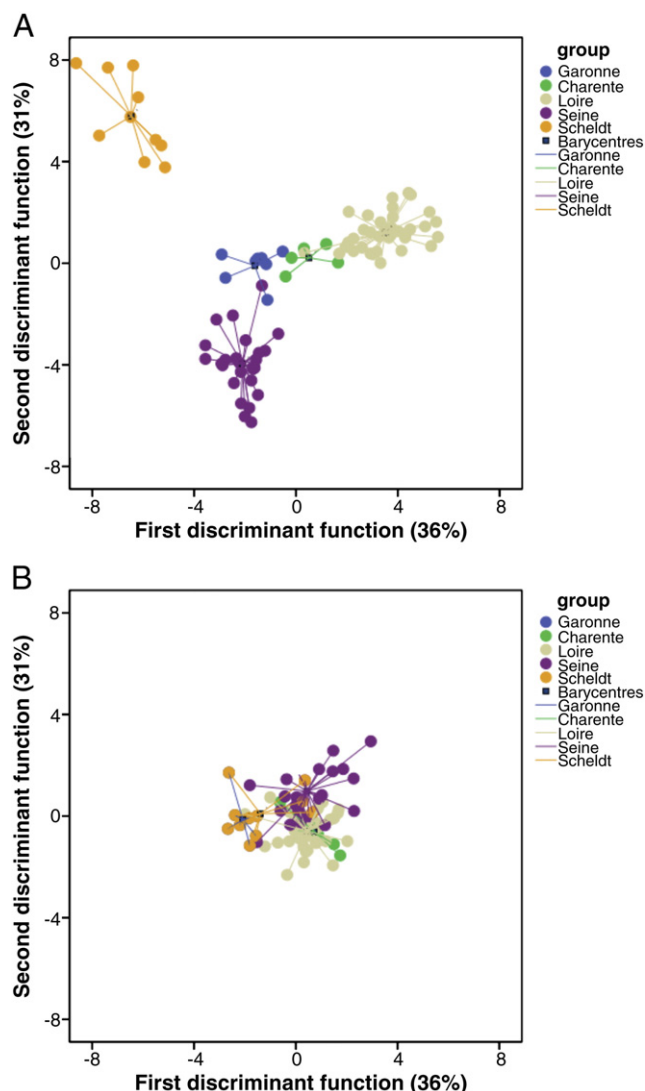


Fig. 1. Plot of the results of discriminant analysis for intersite comparison of contamination patterns on the basis of dry weight normalised trace elements concentrations (A) and of lipid weight normalised organochlorine compound concentrations (B).

et al., 1996). Fish lipid contents can substantially influence the bioaccumulation of organochlorinated compounds (Loizeau, 2001). The organochlorine pollution of the sampled areas is attributable to many sources of industrial, urban and agricultural contamination. The sea bass sampled near the river Scheldt showed the highest levels of total PCBs and DDTs (lw). These results are in good agreement with the French pollution monitoring mussel watch programme which shows that rivers are the main input of pollutants in a local coastal ecosystem, with high levels in regions near industrial or urban areas and that the Seine estuary is one of the most PCB contaminated in Europe (Abarnou, 2003). Catchment areas of the rivers are one of the driving factors explaining OCs variability in European sea bass. Indeed, the Scheldt catchment area (20,000 km²) shelters more than 50% of the total Belgian surface and covers a very densely populated and highly industrialised area of northern France, western Belgium and the south western Netherlands (Voorspoels et al., 2004/9). A lower total PCB and total DDT contamination has been observed in sea bass sampled in the coastal regions near the two longest rivers of France, Loire and Seine. The catchment area of the Loire (117,000 km²) represents 20% of the whole French surface while the Seine catchment area (78,600 km²) shelters approximately 18 million inhabitants and encompasses 40% of the French industrial activity as well as significant agricultural activity (Abarnou, 2003; Abarnou et al., 2002; Bodin et al., 2007; Loizeau et al.,

2001). The lowest levels of total PCB and total DDT were observed in the sea bass sampled in the coastal regions near the Charente and the Gironde. These rivers are much shorter and have smaller catchment areas (Durrieu et al., 2005) (Table 2).

The concentrations of ICES PCBs and DDTs measured in the muscles of European sea bass are reviewed and compared with those found in similar wild species from other regions. It can be seen that the mean levels of ICES PCBs detected in specimens from the French and Belgian coast are generally higher than those reported from other Atlantic and Mediterranean areas. Otherwise, the DDT levels were comparable to those from Italy and Portugal (Antunes and Gil, 2004; Carubelli et al., 2007; Trocino et al., 2007) but substantially lower than those from Spain and Greece (Pastor et al., 1996; Schnitzler et al., 2008).

4.2. Contaminant profile information

The contamination pattern differences of organochlorine pollutants among the sampling sites revealed by the discriminant analysis can be visualised by several charts showing the PCB pattern fractions, the fractioning of the different analysed pesticides as well as the fractioning of the DDT metabolites (Fig. 2).

The levels of organochlorines were as follows PCBs > DDTs > aldrin and dieldrin > HCHs. Although there are differences among sampling regions, PCB patterns were always dominated by a large contribution from the hepta-, hexa- and pentachlorinated PCBs, which collectively accounted for 81 to 86% of the PCBs. These congeners are the most abundant due to their common use in commercial mixtures such as Aroclor 1254 and 1260, but also to their high lipophilicity, stability and persistence that facilitate the adsorption to sediments and the accumulation in the aquatic system, and to their molecular structure (Naso et al., 2005). The tri-, tetra and octachlorinated PCBs were detected at noticeably lower concentrations (Fig. 2A).

Despite the considerable length of time that has passed since 1972 when legal restrictions were introduced for the use of DDT in many European countries, DDT and his metabolites have been detected in all the samples (Fig. 2B). In most of the cases *pp'*DDD was the principal form of DDTs, constituting over 50% of the DDTs. The *pp'*DDD form was dominant in sea bass muscle from the coastal regions near the Loire, Seine and Scheldt. The *pp'*DDT was the less represented form in all the groups constituting less than 20% of the DDTs. The *pp'*DDE constitutes around 30% of the DDT metabolites but is the dominant form in sea bass muscle from the coastal regions near the Gironde and the Charente. The DDT metabolites (*pp'*DDE and *pp'*DDD/*pp'*DDT) ratio is commonly used to assess the chronology of DDT input into the ecosystems (Bordajandi et al., 2003). The ratio greater than 1 suggests that there has been no recent input of DDT in these regions (Fig. 2C).

The situation for the organochlorine pesticide aldrin and his degradation product dieldrin may also give an indication of the chronology of aldrin input in the ecosystem (Gonzalez et al., 2003). Dieldrin was the dominant form measured in the muscles of sea bass in all sampling locations, the dieldrin/aldrin ratio in these cases were higher than 1, indicating no recent input of aldrin in these regions.

The relatively low HCH concentrations might be attributed to the fact that, in contrast to PCBs and DDTs, HCHs are not magnified through the food web (Pastor et al., 1996). The observed concentrations may be ascribed to its previous use as insecticides and fumigants on a wide range of soil-dwelling and plant-eating insects, as well as in personal care products for the control of lice and mites, but also the fact that it is a by-product in the manufacturing processes of various chlorine-containing chemicals and an impurity in several pesticides (Bailey, 2001; Brown et al., 2004).

4.3. Risk assessment

Persistent organochlorinated compounds have a strong tendency to accumulate in lipids, and as a consequence, concentrations are usually

Table 2

Mean contamination levels of zinc (Zn), selenium (Se), copper (Cu), manganese (Mn), mercury (Hg), nickel (Ni), lead (Pb) and cadmium (Cd) in the white muscle of European sea bass collected from coastal regions near several important European river mouths. The concentrations are given in $\mu\text{g kg}^{-1}$ wet weight and dry weight (mean \pm standard deviation, (median) and min–max) * significant ANOVA; ^{a,b,c} significant after Tukey's multiple comparisons.

Coastal region	Gironde	Charente	Loire	Seine	Scheldt	ANOVA
n	8	8	34	26	11	
<i>Wet weight</i>						
Zn ($\mu\text{g kg}^{-1}$)	5330 \pm 750 (5250) 4450–6940	3910 \pm 1760 (3280) 2720–7390	3970 \pm 760 (3790) 2950–5780	5180 \pm 2200 (4710) 3150–1510	5370 \pm 760 (5370) 4830–5900	$F(4,83) = 3.8$ $p = 0.008^*$
Se ($\mu\text{g kg}^{-1}$)	470 \pm 30 ^a (460) 420–510	310 \pm 80 ^b (290) 240–470	350 \pm 60 ^{b,c} (340) 240–590	330 \pm 80 ^b (320) 200–570	790 \pm 540 ^{a,c} (790) 410–1170	$F(4,83) = 14.2$ $p < 0.001^*$
Cu ($\mu\text{g kg}^{-1}$)	350 \pm 80 (340) 260–530	380 \pm 300 (270) 160–720	280 \pm 120 (260) 160–980	290 \pm 110 (280) 130–560	410 \pm 60 (410) 370–450	$F(4,83) = 1.1$ $p = 0.384$
Mn ($\mu\text{g kg}^{-1}$)	280 \pm 90 (270) 160–420	130 \pm 50 (130) 70–220	220 \pm 160 (140) 80–620	210 \pm 110 (190) 70–490	220 \pm 60 (220) 170–620	$F(4,83) = 1.4$ $p = 0.244$
Hg ($\mu\text{g kg}^{-1}$)	90 \pm 30 ^a (90) 60–160	180 \pm 60 ^{b,c} (170) 110–250	150 \pm 40 ^{a,b,c} (150) 60–250	210 \pm 80 ^c (190) 120–440	130 \pm 20 ^{a,b} (130) 110–140	$F(4,83) = 7.9$ $p < 0.001^*$
Ni ($\mu\text{g kg}^{-1}$)	80 \pm 30 (70) 40–110	60 \pm 30 (50) 30–120	100 \pm 60 (80) 30–330	70 \pm 80 (50) 30–430	60 \pm 30 (60) 40–80	$F(4,83) = 1.3$ $p = 0.273$
Pb ($\mu\text{g kg}^{-1}$)	20 \pm 10 ^a (20) 10–30	30 \pm 40 ^{a,b} (20) 10–110	50 \pm 20 ^b (40) 10–120	40 \pm 30 ^{a,b} (30) 10–120	60 \pm 20 ^{a,b} (60) 40–70	$F(4,83) = 2.6$ $p = 0.041^*$
Cd ($\mu\text{g kg}^{-1}$)	1 \pm 4 ^a (1) 1–10	8 \pm 13 ^b (1) 1–30	3 \pm 7 ^{a,b} (1) 1–30	1 \pm 3 ^a (1) 1–10	1 \pm 1 ^a (1) 1–2	$F(4,83) = 2.5$ $p = 0.047^*$
<i>Dry weight</i>						
Zn ($\mu\text{g kg}^{-1}$)	24,200 \pm 3420 (23,900) 20,200–31,500	17,800 \pm 8010 (14,900) 12,400–33,600	18,000 \pm 3460 (17,200) 13,400–26,300	23,500 \pm 9990 (21,400) 14,300–68,600	24,400 \pm 3420 (24,400) 22,000–26,800	$F(4,83) = 3.8$ $p = 0.010$
Se ($\mu\text{g kg}^{-1}$)	2130 \pm 130 ^a (2100) 1890–2300	1420 \pm 370 ^b (1330) 1110–2150	1580 \pm 290 ^{a,b} (1560) 1110–2660	1520 \pm 370 ^{a,b} (1420) 920–2600	3590 \pm 2440 ^c (3590) 1860–5310	$F(4,83) = 14.1$ $p < 0.001^*$
Cu ($\mu\text{g kg}^{-1}$)	1600 \pm 370 (1520) 1180–2390	1710 \pm 1370 (1230) 730–4450	1290 \pm 540 (1190) 750–3290	1330 \pm 480 (1270) 600–2550	1880 \pm 250 (1880) 1700–2060	$F(4,83) = 1.1$ $p = 0.279$
Mn ($\mu\text{g kg}^{-1}$)	1260 \pm 410 (1190) 720–1890	590 \pm 240 (570) 330–1000	980 \pm 710 (660) 370–2830	960 \pm 500 (850) 330–2210	990 \pm 280 (990) 790–1190	$F(4,83) = 1.3$ $p = 0.368$
Hg ($\mu\text{g kg}^{-1}$)	420 \pm 140 ^a (410) 280–720	800 \pm 280 ^{a,b} (750) 520–1140	680 \pm 200 ^{a,b} (660) 250–1140	950 \pm 350 ^b (860) 520–2000	570 \pm 90 ^{a,b} (570) 500–630	$F(4,83) = 8.0$ $p < 0.001^*$
Ni ($\mu\text{g kg}^{-1}$)	330 \pm 110 (320) 190–480	260 \pm 140 (220) 160–520	440 \pm 260 (370) 150–1510	320 \pm 350 (230) 140–1970	280 \pm 110 (280) 200–350	$F(4,83) = 1.3$ $p = 0.309$
Pb ($\mu\text{g kg}^{-1}$)	70 \pm 40 (70) 30–150	140 \pm 170 (70) 50–490	210 \pm 100 (200) 70–540	180 \pm 150 (120) 40–570	260 \pm 80 (260) 200–310	$F(4,83) = 2.6$ $p = 0.052$
Cd ($\mu\text{g kg}^{-1}$)	20 \pm 10 (10) 10–40	40 \pm 50 (10) 10–130	20 \pm 20 (20) 10–130	10 \pm 10 (10) 10–40	20 \pm 10 (20) 10–20	$F(4,83) = 2.3$ $p = 0.073$

normalised on lipid basis. However, if the purpose is to investigate the level of contamination in order to assess human intake, data on fresh weight basis are far more useful. Food consumption is the main exposure route for organochlorinated pollutants for the general population, and fish and fishery products seem to be the main contributors to the total dietary intake of these pollutants (Foran et al., 2005; Gochfeld and Burger, 2005; Hites et al., 2004; Marcotrigiano and Storelli, 2003; Mozaffarian and Rimm, 2006; Sidhu, 2003; Sioen et al., 2008).

The concentrations of HCHs and DDTs found in all analysed samples were well below the Maximum Residue Limits for organochlorine pesticides in some food products of animal origin (FAO, 2008). The 7 ICES PCB levels were below the limit of 75 ng g⁻¹ fw Maximum Residue Limits for muscle meat of fish (A.R. du 06/03/02).

But based on the tolerable daily intake for the seven indicator PCBs of 0.01 $\mu\text{g kg}^{-1}$ set by the AFSSA (2009), the maximum amount of European sea bass flesh that can be eaten by an average adult (60 kg)

before reaching the safety limits was estimated at 680 g (for sea bass from the coastal zone near the Seine) up to 2250 g (for sea bass from the coastal zone near the Charente) per month. Combined with the average fish proportion size of 227 g (Carubelli et al., 2007) this represents 3 to 10 sea bass meals per month. These results may be an important issue for human communities who regularly consume fish, such as coastal populations. The levels in European sea bass are among the highest measured in edible marine fish species, next to eel and trout (Naso et al., 2005). This probably reflects their feeding habits and the nature of the habitat of these benthic and euryhaline species. They usually inhabit shallow waters with sandy or muddy bottoms along the coast, ports and estuaries, which are generally considered to be more heavily polluted than open waters (Lewis et al., 2002; Loizeau, 2001).

Various national scientific bodies formulate the recommendations that the general population should consume fish at least twice a week, including some oily fish, and that pregnant or breast-feeding women

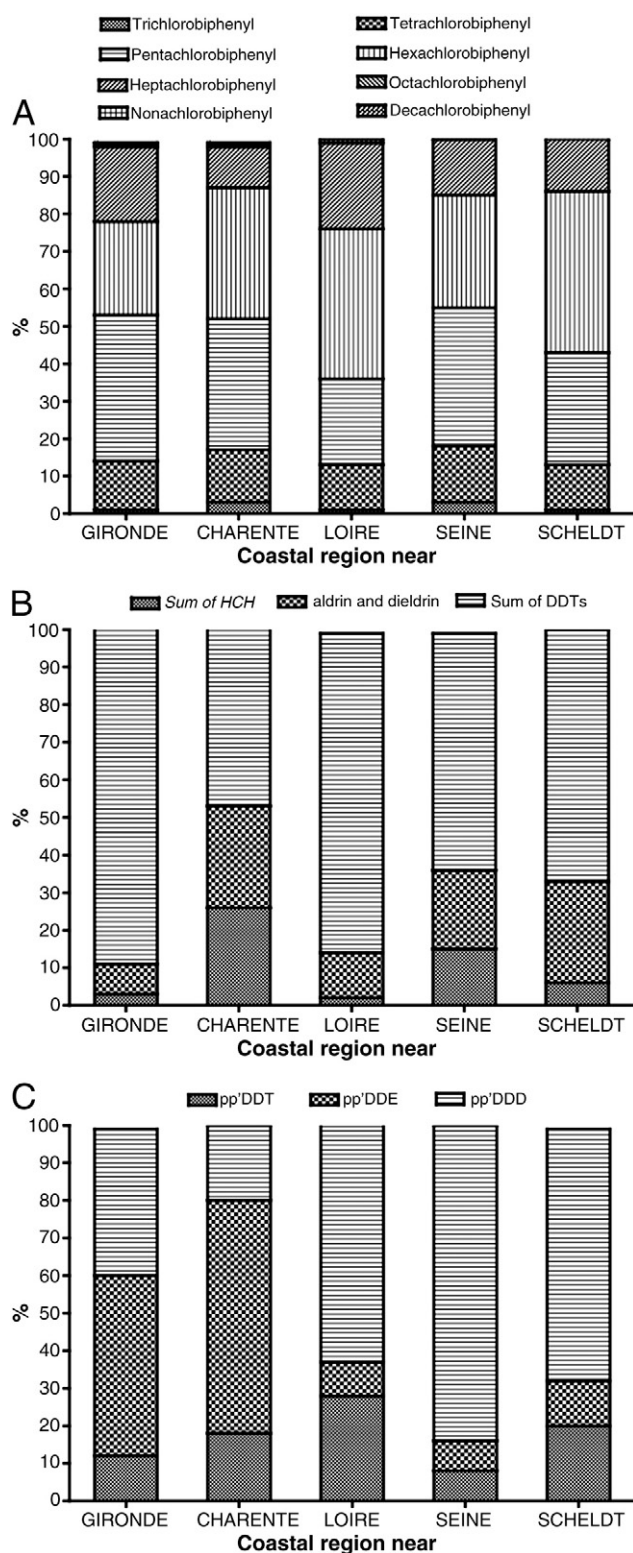


Fig. 2. Geographical differences in contamination patterns PCB congeners (A), chlorinated pesticides (B) and DDT metabolites (C).

should consume predator fish not more than once a week (Hites et al., 2004; Marcotrigiano and Storelli, 2003; Sidhu, 2003). Looking beyond these general recommendations, this study highlights the advantages of diversifying the consumed fish and seafood species in terms of proportions and provisioning origins in order to ensure a rational balance between benefits and risks compatible with nutritional and

toxicological recommendations (Foran et al., 2005; Gochfeld and Burger, 2005; Mozaffarian and Rimm, 2006; Sioen et al., 2008).

4.4. Trace elements

4.4.1. Contamination level

The trace element concentrations measured in our sampling are quite low and are part of the background contamination level that is relatively homogeneous all along the French coast. Generally metal accumulation is the highest in liver and gills, while it is low in gonad and muscle in all fish species (Dural et al., 2006). These organs are also good indicators of chronic exposure to heavy metals because they are the sites of metal metabolism (Dugo et al., 2006). The liver is often considered a good monitor of water pollution with metals since their concentrations are proportional to those present in the environment, whereas muscle is a poor indicator (Dural et al., 2006). In European sea bass, the contamination levels in liver are two to ten times higher than in muscle, although no direct relation was published (Dural et al., 2006). The levels of Zn, Se, Cu, Mn, Hg, Ni, Pb and Cd were determined in the muscle because of its importance for human consumption. Our measured concentrations are mostly in the lower range measured in European sea bass (Dugo et al., 2006; Dural et al., 2006; Durrieu et al., 2005; Miramand et al., 2001) and other fish (Bustamante et al., 2003; Canli and Atli, 2003; Cohen et al., 2001; Kalay et al., 1999; Kwon and Lee, 2001; Topcuoglu et al., 2002; Türkmen et al., 2005; Tüzen, 2003).

4.4.2. Contaminant profile information

Although the concentrations of several trace elements (Zn, Se, Hg, Pb and Cd) differed significantly among sampling locations, the discriminant analysis could not separate the predefined groups of individuals (sampling location of sea bass) by given measurements of trace element variables. There were no systematic differences that would have supported distinct contamination patterns for the different regions.

4.4.3. Risk assessment

The highest Hg levels have been measured in sea bass from coastal regions near the river Seine, but the mean level is more than 4 times lower than the maximum authorised limit of 1 mg/kg (EC, 2005). The highest Pb levels have been measured in sea bass from coastal regions near the river Loire, but the mean level is more than three times lower than the maximum authorised limit of 0.2 mg/kg (EC, 2005). Finally, the highest Cd levels have been measured in sea bass from coastal regions near the river Charente, but the mean level is more than 6 times lower than the maximum authorised limit of 0.05 mg/kg (EC, 2005). The risk linked to metal exposure through European sea bass consumption appeared therefore limited. Based on the tolerable weekly intake for Hg set by the FAO/WHO (5 µg/kg b.w./week) (FAO/WHO, 2003), the maximum amount of European sea bass flesh that can be eaten by an average adult (60 kg) before reaching the safety limits was estimated at 1828 g (for sea bass from the coastal zone near the Seine) up to 4266 g (for sea bass from the coastal zone near the Gironde) per month. Combined with the average fish proportion size of 227 g (Carubelli et al., 2007), this represents 8 to 18 sea bass meals per month. The safety limits for Pb and Cd (FAO/WHO, 1999, 2000) are unreachable; the amount of sea bass flesh that can be eaten by an average adult (60 kg) exceeds the 100 portions per month.

Zinc, Se, Cu and Mn are elements that need to be present in the human diet to maintain normal physiological functions (Goldhaber, 2003). A regular consumption of sea bass may present a source for essential trace elements. A portion sea bass supplies up to 128 to 194% of the RDA of 0.055 mg/day for Se. These levels are 3 to 5 times lower than the tolerable upper intake level of 0.4 mg/day. There is thus no risk of toxicity due to high selenium exposure through sea bass diet. Besides this, a sea bass meal provides up to 8–11% of the Recommended Dietary Allowance (RDA) for Zn and Cu (11 mg/day and 0.9 mg/day respectively) and 2–4% of the RDA of 1.6 mg/day for Mn.

5. Conclusions

The non-essential trace element concentrations measured in our sampling were quite low and the risk linked to metal exposure through European sea bass consumption appeared therefore limited. High contamination levels of organochlorinated compounds were measured in the muscles of European sea bass sampled in the coastal regions near the Scheldt, Seine, Loire, Charente and Gironde. The Scheldt and the Seine are still among the most contaminated estuaries in Europe. These levels were generally higher than those reported in literature of sea bass from other regions. Each region presented their specific contamination patterns reflecting different sources due to the input of the respective rivers. As fish and fishery products are the main contributors of the total dietary intake of organochlorinated pollutants, regular consumption of European sea bass with the reported contamination levels may represent an important exposure route for the general population.

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