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Concentration of polychlorinated biphenyls and organochlorine pesticides in mullet (*Mugil cephalus*) and sea bass (*Dicentrarchus labrax*) from Bizerte Lagoon (Northern Tunisia)

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HIGHLIGHTS

- OCP and PCB levels were reported in fish from Bizerte Lagoon (Tunisia).
- OCs levels were comparable or slightly higher than to those found in other studies.
- The estimated daily intake for DDTs and γ-HCH were far below the ADI recommended by FAO/WHO.

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ABSTRACT

Concentrations of polychlorinated biphenyls (PCBs), and organochlorine pesticides (OCPs) were determined in two fish species, mullet (*Mugil cephalus*) and sea bass (*Dicentrarchus labrax*), collected from Bizerte Lagoon and the Mediterranean Sea. In all samples, PCBs were found in higher concentrations than OCPs. The highest concentrations of OCPs and PCBs were found in sea bass, and in Bizerte Lagoon. Concentrations of DDTs and PCBs detected in this study were generally comparable or slightly higher than those found in studies from other Mediterranean and non-Mediterranean regions subject to a high anthropogenic impact. ∑PCBs, ∑HCHs and HCB levels were negatively correlated with lipid content, while no such correlation was seen for ∑DDTs. A significant correlation between levels and length and between levels and weight existed only for ∑PCBs. The daily intake of PCBs and OCPs ingested by people living in Bizerte through the studied fish species was estimated and compared with those observed in other areas.

1. Introduction

Environmental contamination by organochlorine pesticides (OCPs) and polychlorinated biphenyls (PCBs), which are representative persistent organic pollutants (POPs), is a serious problem for human beings. OCPs and PCBs have been produced and used for agricultural and industrial purposes for a long time and on a large scale. PCBs were used in a wide range of applications, such as coatings, inks, flame retardants, paints, heat-transfer systems, electrical transformers and hydraulic fluids. Organochlorine pesticides had been used throughout the world thanks to exceptional insecticidal and fungicidal properties (Xu et al., 2010). Following the extensive research concerning their wide distribution in the environment and toxic effects on human and animal health had been discovered, their production and use have been restricted and/or banned. Because of their high chemical stability, long-range atmospheric transport, lipophilicity and persistence, these chemicals tend to bioconcentrate and biomagnify in the food chains and persist in the environment for many years, representing a definite health hazard for both wildlife and humans (Naso et al., 2005; Covaci et al., 2006; Roche et al., 2009). Contamination by PCBs and OCPs has been reported in air, water, sediment, soil, vegetables, fish, meats and in human sample, like breast milk, blood, and adipose tissue. Long-term chronic exposure to OCPs and PCBs has been correlated with severe injury to the nervous, endocrine, reproductive, and immune systems in birds, fish, and mammals (Ratcliffe, 1967; Kelce et al., 1995; Skaare et al., 2000; Toft et al., 2003). Ingestion is the main source of human exposure to organochlorine pollutants, and, in particular, the consumption of seafood from contaminated

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areas (Alcock et al., 1998). Moreover, several studies have recently demonstrated a clear correlation between the frequency of fish consumption and the levels of organochlorines in human tissues, serum, and milk (Kiviranta et al., 2002; Fitzgerald et al., 2004). POPs in fish from some areas were detected to assess the risk for human health (Crane, 1996; Binelli and Provini, 2004; Yang et al., 2006).

Organochlorine compounds (OCs) including OCPs and PCBs were widely used in Tunisia from the end of 1940s to 1980s. OCPs were used for insect control, sanitary purposes, and agricultural and building protection. PCBs have been widely employed in industry as heat exchange fluids, in electric transformers and capacitors. Despite the use of these compounds has been prohibited, they have also been found in sediments (Cheikh et al., 2002; Derouiche et al., 2004; Ameur et al., 2011), in biota (Covaci et al., 2006; de Mora et al., 2010), in human blood, breast milk and serum (Ennaceur et al., 2008; Ennaceur and Driss, 2010).

Bizerte Lagoon (Northern Tunisia) is an area of vital environmental importance; many resident marine species live, feed, and die in this area, and many of the pelagic species reproduce there. Among them, there is thirty fish species (Harzallah, 2003). However, this lagoon is submitted to many anthropic pressures including urbanization, industrial activities (cement works, metallurgical industry, boatyard, tyre production factories, etc.), as well as naval and commercial shipping harbors. Lagoon shores have also been used as open-air waste-dumping sites. The direct and indirect discharges of urban and industrial wastes and runoff lead to the chemical contamination of the lagoon by various toxic compounds such as OCPs (Cheikh et al., 2002), halogenated aromatics compounds like PCBs (Derouiche et al., 2004), polycyclic aromatic hydrocarbons (PAHs) (Trabelsi and Driss, 2005), heavy metals (Ben Garali et al., 2010) and organotins (Abidli et al., 2011). Despite the presence of all these possible contaminants, there are no studies about the accumulation of persistent halogenated pollutants in marine species from the Bizerte Lagoon.

To acquire further data on the state of contamination of Bizerte Lagoon and to assess potential risks for fish consumers, this study investigated the residue levels of persistent organochlorine pollutants in two fish species among its edible marine species. Both species have high economic importance and gastronomic value, which means that the information provided by our study may be important both to people using the lagoon resources and to those responsible for fishery management. Moreover, given the existence of a positive correlation between fish consumption and Tunisian breast milk levels of pesticides and PCBs (Ennaceur et al., 2008) and the relatively high consumption of fish in Tunisia (average consumption of fish is 9 kg per person per year according to the National Institute of Nutrition and Food Technology), it is necessary to clarify the status of their contamination in such foodstuffs.

2. Materials and methods

2.1. Sample collection

Bizerte Lagoon is a lagoon, located in Northern Tunisia. It extends for about 150 km^2, between latitude 37°08 and 37°14 N and longitude 9°48 and 9°56E and is connected to the Mediterranean Sea and Lake Ichkeul by straight channels. The Mediterranean Sea far from the main polluting sources was selected as a reference site.

The studied fish species collected in the present study included mullet (*Mugil cephalus*), and sea bass (*Dicentrarchus labrax*). Fifteen fish samples from Bizerte Lagoon and five fish samples from the Mediterranean Sea, for each of both analyzed species, were sampled using a net, in December 2010 (Fig. 1). The fish were immediately sacrificed, weighed, measured, dissected and kept frozen (−20 °C) until required for chemical analyses. Table 1 shows the information on the fish samples used in the present study.

2.2. Chemicals

The solvents used in this study (n-hexane, acetone and dichloromethane) were pesticide quality and were obtained from Fluka (Buchs, Switzerland). Sulfuric acid was obtained from Biotechnica. Florisil (60–100 mesh) was obtained from Merck (Darmstard, Germany), activated at 650 °C for 8 h and and re-heated at 130 °C for 5 h before use. Thereafter, it was stored in a desiccator until use. Anhydrous sodium sulfate suitable for use in pesticide analysis was purchased from Fluka, heated at 300 °C and stored in a 130 °C oven.

OCPs standards including p,p′-DDE, p,p′-DDD, o,p′-DDD and p,p′-DDT (the sum expressed as ∑DDTs), HCB, β-HCH and γ-HCH (the sum expressed as ∑HCHs) with purities ranged from 97% to 99%, were obtained from Polyscience Corporation Analytical Standards (Niles, IL, USA) and individual standard solution of each pesticide were prepared in hexane at 1000 μg mL^{-1} except β-HCH which is dissolved in acetone. A standard mixture of twelve PCBs congener (PCB-18, 28, 31, 52, 44, 101, 149, 118, 153, 138, 180 and 194) (the sum expressed as ∑PCBs) at 10 μg mL^{-1} in heptane was purchased from Supelco (CIL, USA). These standard solutions were further diluted by n-hexane to obtain mixed fortifying and GC calibration standard solutions for all organochlorines.

2.3. Sample preparation and extraction

OCPs and PCBs were analyzed in fish muscles following the method described by Guo et al. (2008) with slight modifications. Freeze dried muscle tissue (10 g) was soxhlet extracted with n-hexane:acetone (4:1; v/v) for 16 h a rate of five cycles per hour. The extract was concentrated with a rotary evaporator. An aliquot of 1 mL was used for gravimetric determination of the extractible lipid content. The remaining lipids were removed by treatment with concentrated sulfuric acid (4× 10 mL). Further, cleanup was done on a column (40 cm × 0.5 cm ID) packed with 5 g of activated florisil and topped with 1 g of anhydrous sodium sulfate. The extract was eluted with 50 mL of dichloromethane and n-hexane (1:9; v/v). The eluate was finally concentrated in a Kuderna–Danish to 1 mL and was ready for Gas Chromatography (GC) analysis.

2.4. GC analysis

The analysis of OCPs and PCBs was performed on an Agilent Model 6890 Gas Chromatograph equipped with a ^{63}Ni electron capture detector (GC–ECD) and operated by HP Chemstation Software. In total, 2 μL of extract was injected in splitless mode into a PTE-5 (30 m × 0.32 mm i.d., 0.32 μm film thickness) capillary column, using hydrogen (H_2) carrier gas with a 1 mL min^{-1} flow rate and the following oven temperature program: 50 °C initial (2 min) to 160 °C at 5 °C min^{-1} and to 260 °C at 2 °C min^{-1}, hold 10 min. The temperatures of the injector and detector were 250 and 300 °C, respectively. Quantitative and qualitative analysis of OCPs and PCBs were done by comparison with external standard. Compounds analyzed by GC–ECD were confirmed in each extract sample by GC–MS using a Finningan Trace MS instrument, working at electron impact ionization mode and using splitless injection mode and helium as carrier gas. GC–MS was operated by Xcalibur Software and was equipped with a DB5-MS capillary column (30 m × 0.25 mm i.d., 0.25 mm film thickness).

The oven temperature program used for the analysis of PCBs was the following: 60 °C (hold for 1 min), increased at 8 °C min^{-1} to 175 (hold for 1 min), at 3 °C min^{-1} to 205 °C (hold for 5 min),

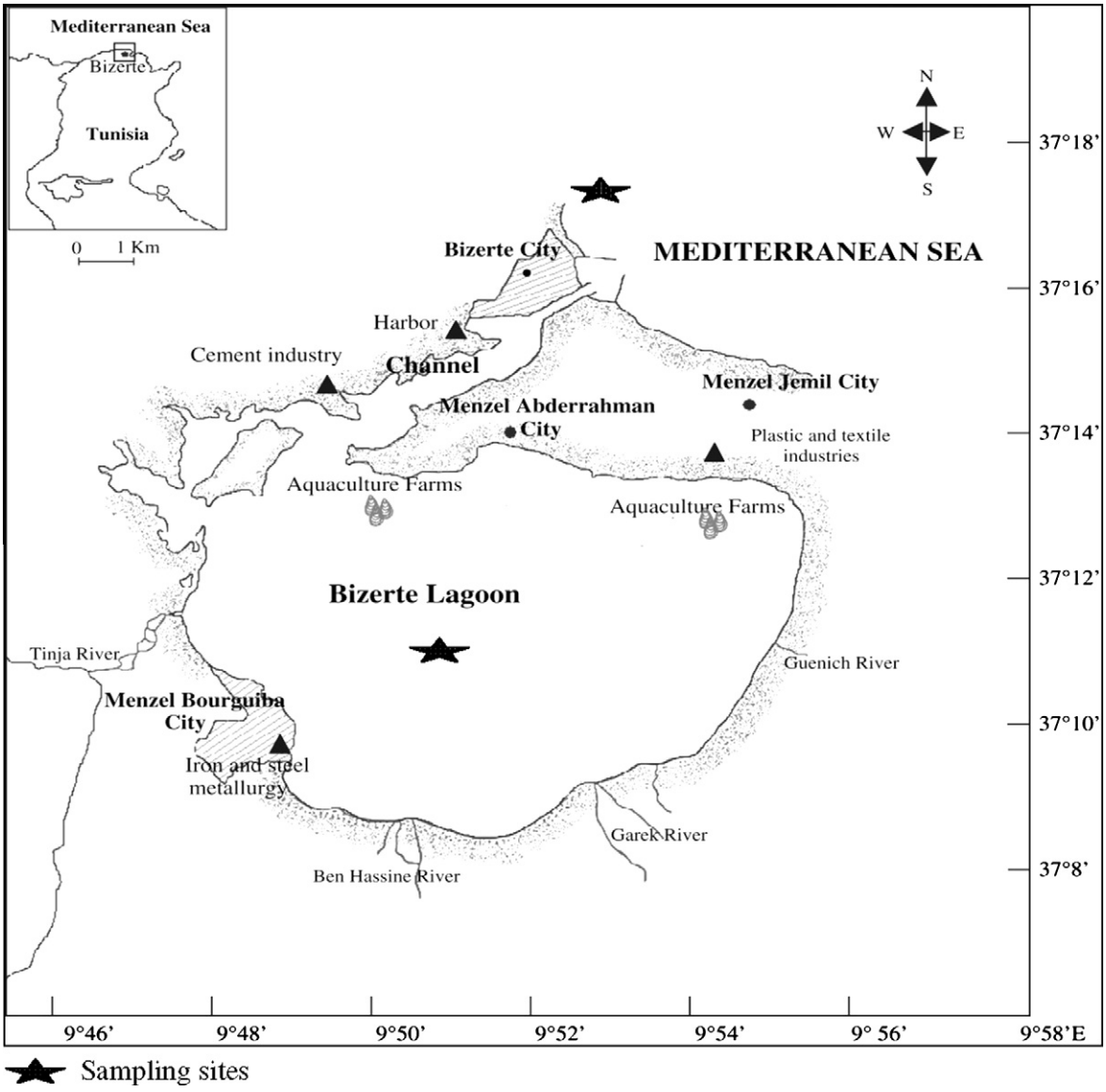

Fig. 1. Map showing sampling areas.

at 10 °C min^{-1} to 300 °C (hold for 5 min) and at 10 °C min^{-1} to 320 °C (hold for 2 min). The oven temperature program used for the analysis of OCPs was the following: 60 °C (hold for 1 min), increased at 8 °C min^{-1} to 175 (hold for 1 min), at 3 °C min^{-1} to 205 °C (hold for 5 min), and at 10 °C min^{-1} to 300 °C (hold for 15 min). The mass spectrometer was run with an EI+ source, with a source temperature of 200 °C, interface temperature of 280 °C and electron energy of 70 eV. Compounds were determined by selected ion monitoring (SIM). The ions monitored were: 256, 258 (tri-CB); 290, 292 (tetra-CB); 326, 354 (penta-CB); 360, 290 (hexa-CB); 394, 324 (hepta-CB); 430, 179 (octa-CB); 181, 183 (β-, γ-HCH); 284, 286 (HCB); 246, 248 (p,p′-DDE); 235, 237 (o,p′-DDD, p,p′-DDD, o,p′-DDT, p,p′-DDT).

2.5. Quality control

Procedural blanks were analyzed simultaneously with every batch of five samples to check for interferences or contamination from solvent and glassware. No analytes of interest were detected. Using the described methodology, recovery done at 10 ng mL^{-1} and 40 ng mL^{-1} levels, ranged from 70% to 90%, for OCPs and from 60% to 82% for individual PCB congeners. All samples were recovery corrected. Relative standard deviations of the method ($n = 5$) were below 10%, indicating acceptable repeatability of the method. The correlation coefficients of the calibration curves were superior to 0.995 across a concentration range of 2.5–25 ng mL^{-1}, representing good linearity of the method. The limits of detection (LOD) calculated by a signal-to-noise ratio of 3, were in the range of 0.5–1 ng g^{-1} lipid wt for OCPs and PCBs.

2.6. Statistical analysis

Statistical treatment of the obtained results was performed with SPSS software (SPSS 10.0 for Windows, SPSS Inc.). Our data were not normally distributed. In the first step we tried to log-transform the data, but even then most of them were still not normally distributed. Therefore we further used non-parametric tests for these data. For general comparisons, we used *k*-independent sample and if difference was significant ($p < 0.05$), subsequent multiple comparisons between fish groups and sites were tested using Mann–Whitney *U*-test. Spearman rank correlation was used

Table 1
Details of samples from Bizerte Lagoon (BL) and the Mediterranean Sea (MS).

Common name	Scientific name	Number of samples		Weight (g) (±SD)		Length (cm) (±SD)		Lipid (%) (±SD)		Feeding habits	Habitat
		BL	MS	BL	MS	BL	MS	BL	MS		
Mullet	*Mugil cephalus*	15	5 (83.4)	266	185 (15.0)	30.5 (3.50)	26.7 (1.72)	6.27 (5.40)	5.73 (1.62)	Omnivorous (Zooplankton, benthic organisms, detritus, diatom algae, and invertebrates)	Sandy and muddy bottoms
Sea bass	*Dicentrarchus labrax*	15	5	265 (52.6)	290 (10.0)	29.4 (1.31)	30.7 (1.01)	4.56 (3.13)	6.07 (4.37)	Carnivorous (Fishes, crustaceans and molluscs)	Muddy and slimy bottoms

SD: Standard deviation.

to examine the strength of associations between parameters. Statistical significance was accepted at $p < 0.05$.

3. Results and discussion

The concentrations of OCs expressed in nanograms per gram lipid weight (ng g^{-1} lw) in the muscle of the analyzed species are presented in Table 2. OCPs and PCBs were detected in all samples, indicating ubiquitous contamination by these compounds in the studied fish species from Bizerte Lagoon. In general, the residue pattern of OCs in the studied fish species followed the order of PCBs > DDTs > HCHs > HCB. Our results were in accordance with those reported in fish samples from Italy (Jiang et al., 2005) and from China (Naso et al., 2005).

The concentrations of the studied compounds in fish samples varied with the species in both investigated areas. In Bizerte Lagoon, the sea bass had the maximum levels of ∑(OCPs + PCBs) (667 ng g^{-1} lw), while the mullet had the lower levels of ∑(OCPs + PCBs) (357 ng g^{-1} lw). In the Mediterranean Sea, the levels of ∑(OCPs + PCBs) were 252 and 130 ng g^{-1} lw respectively in the sea bass and the mullet (Table 2). A statistically significant differences in ∑(OCPs + PCBs) concentration existed between mullet and sea bass ($p < 0.05$). On the other hand and comparing the two sampling sites, ∑(OCPs + PCBs) in the two studied species were higher in Bizerte Lagoon than in the Mediterranean Sea (Table 2). A statistically significant difference in concentration was observed between sites ($p < 0.05$).

3.1. Concentration of DDTs in samples

Mean concentrations of ∑DDTs in the studied fish species from Bizerte Lagoon and from the Mediterranean Sea were 134 and 23.4 ng g^{-1} lw respectively. Despite the considerable length of time that has passed since 1980 when legal restrictions were introduced for the use of DDT in Tunisia, its metabolites (p,p′DDE, p,p′DDD and o,p′DDD) were found in all the analyzed samples. p,p′-DDE was present in much higher concentrations than HCB, HCHs, p,p′DDD, o,p′DDD and p,p′DDT (ranging from 23.4 to 114 ng g^{-1} lw and from 5.43 to 17.5 ng g^{-1} lw respectively in the studied fish species caught from Bizerte Lagoon and from the Mediterranean Sea). p,p′-DDD levels ranged from 10.2 to 62.5 ng g^{-1} lw and from 5.45 to 13.5 ng g^{-1} lw respectively in the studied fish species collected from Bizerte Lagoon and from the Mediterranean Sea. o,p′-DDD levels varied from 2.59 to 57.3 ng g^{-1} lw and from 1.66 to 5.98 ng g^{-1} lw respectively in the studied fish species collected from Bizerte Lagoon and from the Mediterranean Sea. p,p′-DDT was only found in both studied fish species collected from Bizerte Lagoon (33.33% and 25% of the samples respectively in mullet and sea bass) at concentrations between nd and 25.6 ng g^{-1} lw.

DDT and its derivatives followed this general order in this study: DDE > DDD > DDT. Our results are in accordance with reported order of metabolites of DDT in fish from other countries (Perugini et al., 2004; Naso et al., 2005; Covaci et al., 2006; Hosseini et al., 2008; Davodi et al., 2011). DDT is degraded into DDD under anaerobic conditions and into DDE under aerobic conditions (Hitch and Day, 1992). Higher proportions of p,p′-DDE to p,p′-DDD in the analyzed samples suggested that DDT endured aerobic transformin organisms (Zhou et al., 2008). The DDE/DDT ratio is commonly used to assess the chronology of DDT input into the ecosystems. In general, a ratio of (DDE + DDD)/∑DDTs of more than 0.5 indicates long-term biotransformation of DDT to DDE and DDD, while a ratio of less than 0.5 may indicate recent input of DDT (Hitch and Day, 1992). The ratios of (DDE + DDD)/∑DDTs was high in this study, (mean 0.93 and 0.98) respectively in mullet and sea bass from Bizerte Lagoon. While in the Mediterranean Sea, mean of this ratio was 1 for both species. This result indicated aged DDT pollution and suggested that there has been no recent input of technical DDT from the agricultural areas into Bizerte Lagoon.

In Table 3, the concentrations of DDTs detected in the studied fish species from the Bizerte Lagoon are reviewed and compared with those found in similar studies from other Mediterranean and non-Mediterranean regions. It can be seen that the mean levels of DDTs detected in specimens from Bizerte Lagoon are generally comparable to that reported for specimens from the Ria de Aveiro in Portugal (Antunes and Gil, 2004). Concentrations of DDTs were lower than those detected in fishes from other Mediterranean aquatic ecosystems such as the Marmara Sea in Turkey (Coelhan et al., 2006), the Gulf of Naples in Italy (Naso et al., 2005) and the Ebro Delta in Spain (Pastor et al., 1996). Otherwise, the DDT levels found in this study are generally higher than those reported from the other regions such as Iran (Davodi et al., 2011) and USA (Sajwan et al., 2008).

3.2. Concentration of HCHs in samples

In comparison to DDTs, HCHs had lower concentrations (Table 2). In Bizerte Lagoon, β-HCH, and γ-HCH were found in 50% and 96% of samples respectively. Whereas in the Mediterranean Sea they were found in 33.4% and 100% of samples respectively. Total HCHs concentrations in samples collected from Bizerte Lagoon ranged from 0.57 to 20.1 ng g^{-1} lw and from 2.69 to 33.6 ng g^{-1} lw respectively in mullet and sea bass. In the Mediterranean Sea, these concentrations ranged from 1.22 to 2.51 ng g^{-1} lw and from 5.38 to 16.8 ng g^{-1} lw respectively in mullet and sea bass (Table 2). In Bizerte Lagoon, the average concentrations of β-HCH, and γ-HCH contributed 15.4–23.1% (mean 19.3%) and 76.9–84.6% (mean 80.8%), respectively, to the total HCH concentration. The average levels of β-HCH, and γ-HCH contributed 13.7–21.7% (mean 17.7%) and 78.3–86.3% (mean 82.3%), respectively, to the total HCH concentration in the Mediterranean Sea. γ-HCH was the dominant HCH isomer in our samples and coincided with results from other studies (Masmoudi et al., 2001; Zhou et al., 2008).

The technical mixture of HCH usually contains 55–80% α-HCH, 5–15% β-HCH, 8–15% γ-HCH, and 2–16% δ-HCH (Willett et al., 1998). It was evident that the proportions of HCH isomers in the

studied fish species did not reflect the technical mixture composition. Considering also the persistence order of these isomers ($\beta > \gamma > \alpha$; Kouras et al., 1998), this result serves as evidence of the preferential usage of pure γ-HCH (the most toxicological active HCH isomer) than the technical mixture of HCHs on Bizerte Lagoon shores farms.

The concentration of HCHs in the studied fish species in the present study was lower than that of fish from Mediterranean regions such as Italy (Naso et al., 2005) and Turkey (Coelhan et al., 2006) and close to that of fish from the Adriatic Sea (Stefanelli et al., 2004) (Table 3).

3.3. Levels of HCB in specimen

Although the usage of HCB has been banned, all analyzed fish samples contained HCB residues. Mean HCB concentrations were 6.21 ng g^{-1} lw and 10.5 ng g^{-1} lw respectively in mullet and sea bass caught from Bizerte Lagoon. In the Mediterranean Sea, the mean concentrations of this compound were 4.10 ng g^{-1} lw and 12.0 ng g^{-1} lw respectively in mullet and sea bass (Table 2). The present finding of HCB in marine organisms from Bizerte Lagoon may be ascribed not only to its previous use as fungicide treatment for seeds but also to the fact that it can be released from high temperature industrial processes. It may also be present as impurity in other OCPs (Erdogrul et al., 2005). Incineration may also contribute to HCB pollution. The comparison with worldwide studies shows that HCB levels in Bizerte Lagoon fish were lower to those from Spain (Pastor et al., 1996), similar to those from Italy (Stefanelli et al., 2004) and Turkey (Coelhan et al., 2006) and higher than the corresponding levels from others countries such as Iran and USA (Davodi et al., 2011; Sajwan et al., 2008) (Table 3).

3.4. Concentration of PCBs in samples

PCBs were found in both studied fish species (100% of the samples). Residue levels of total PCBs ranged from 164 to 336 ng g^{-1} lw and from 282 to 642 ng g^{-1} lw respectively in mullet and sea bass from Bizerte Lagoon. The total concentrations of PCB residues in fish from the Mediterranean Sea were between 95.3–123 ng g^{-1} lw and between 189–209 ng g^{-1} lw respectively in mullet and sea bass (Table 2). Individual PCB congener distribution patterns were shown in Fig. 2. PCB-118, -138, -153 and -180 were dominant contaminants in the studied fish species collected from both areas, accounting respectively for 9.00%, 14.0%, 28.5% and 23.6% of total PCBs detected in the studied fish species collected from Bizerte Lagoon, while in the studied fish species caught from the Mediterranean Sea, these congeners accounted for 8.10%, 6.62%, 35.1% and 31.2% respectively. Similar results have been reported for PCB-118, -138, -153 and -180 as the predominant congeners in fish from other regions (Naso et al., 2005; Nie et al., 2005).

Profiles of polychlorinated biphenyl congeners, according to the degree of chlorination were shown in Fig. 3. In Bizerte Lagoon, hexachlorobiphenyls were predominant in both caught fish species accounting for 46.3% and 55.0%, respectively in mullet and sea bass, followed by heptachlorobiphenyls with contributions of 24.7% and 22.8% respectively in mullet and sea bass, pentachlorobiphenyls (mullet, 18.7%; sea bass, 17.7) and tetrachlorobiphenyls (mullet, 5.89%; sea bass, 4.52%), while tri-(mullet, 0.66%; sea bass, 0.31) and octachlorobiphenyls (mullet, 3.85%; sea bass, 4.11%) made up a small percentage of the total PCB residues (Fig. 3). Same profiles were observed in the studied fish species from the Mediterranean Sea (Fig. 2). Concerning PCB isomer profile, no discrepancies between fish species was observed. This result is in accordance with that obtained by Davodi et al. (2011). The observed trends in PCB congener compositions were in accordance with those observed in Bizerte Lagoon sediments in Tunisia (Derouiche et al., 2004) and in other studies on fish (Naso et al., 2005; Storelli and Perrone, 2010).

Among the twelve *dioxin-like PCB congeners*, only the PCB 118 was analyzed in the studied fish species. The TEQ values of PCB 118 were calculated using the Toxic Equivalency Factors (TEFs) for fish developed by Van den Berg et al. (1998). Toxic Equivalency

Table 2
OCPs and PCBs concentrations (ng g^{-1} lw) in fishes from Bizerte Lagoon (BL) and the Mediterranean Sea (MS) obtained by GC–ECD analysis.

Compound	*Mugil cephalus*				*Dicentrarchus labrax*			
	BL		MS		BL		MS	
	Mean (SD)	Range	Mean (SD)	Range	Mean (SD)	Range	Mean (SD)	Range
HCB	6.21 (4.58)	1.27–15.1	4.10 (0.26)	3.88–4.39	10.5 (8.32)	1.62–28.5	12.01 (1.33)	10.8–13.5
β-HCH	1.54 (4.09)	nd–14.4	0.25 (0.43)	nd–0.74	2.70 (3.29)	nd–10.1	2.30 (2.19)	nd–4.35
γ-HCH	5.11 (4.58)	nd–15.0	1.56 (0.29)	1.22–1.69	14.9 (10.3)	2.69–30.5	8.33 (3.65)	5.38–12.4
p,p′-DDE	49.8 (16.8)	23.4–78.5	6.23 (0.71)	5.43–6.79	85.0 (14.0)	60.3–114	14.7 (2.68)	12.1–17.5
o,p′-DDD	17.9 (13.4)	2.59–45.6	2.73 (1.01)	1.66–3.67	47.1 (10.4)	20.6–57.3	4.50 (1.76)	2.56–5.98
p,p′-DDD	20.7 (9.10)	10.2–35.8	6.48 (0.92)	5.45–7.22	37.8 (18.8)	11.8–62.5	12.1 (1.39)	10.7–13.5
p,p′-DDT	6.94 (10.6)	nd–25.6	nd		2.83 (5.55)	nd–16.9	nd	
∑HCHs	6.66 (6.07)	0.57–20.5	1.80 (0.65)	1.22–2.51	17.6 (10.3)	2.69–33.6	10.6 (5.74)	5.38–16.8
∑DDTs	95.4 (30.5)	47.3–138	15.4 (11.0)	14.3–16.1	173 (21.2)	156–227	31.3 (5.79)	25.4–36.9
∑OCPs	108 (33.8)	52.9–157	21.3 (1.65)	19.6–22.8	201 (31.1)	158–265	54.0 (9.93)	47.9–65.4
PCB-18	1.64 (5.02)	nd–17.5	nd	nd	1.43 (3.81)	nd–13.3	1.10 (1.91)	nd–3.30
PCB-28 + 31	nq		nq		nq		nq	
PCB-52	9.19 (9.88)	nd–29.8	2.28 (3.96)	nd–6.85	15.0 (10.7)	nd–37.6	3.55 (3.29)	nd–6.51
PCB-44	5.46 (6.63)	nd–20.5	3.16 (2.95)	nd–5.85	6.07 (5.68)	nd–16.6	6.33 (6.06)	nd–12.1
PCB-101	27.2 (11.8)	8.44–43.1	7.61 (0.85)	6.83–8.51	35.0 (13.6)	17.4–60.6	14.6 (0.91)	14.0–15.6
PCB-118	19.3 (8.61)	7.15–35.8	9.14 (0.38)	8.70–9.39	47.8 (27.0)	9.72–92.3	15.5 (0.98)	14.5–16.4
PCB-149	11.5 (10.8)	nd–31.8	3.03 (2.63)	nd–4.71	33.6 (27.6)	nd–84.1	1.87 (3.24)	nd–5.61
PCB-138	44.9 (29.0)	13.4–96.8	11.6 (1.58)	10.2–13.3	46.4 (13.6)	26.4–74.8	5.21 (1.71)	4.02–7.17
PCB-153	58.8 (12.2)	47.5–92.9	29.6 (5.18)	25.2–35.3	156 (22.8)	111–190	85.5 (9.77)	75.6–95.2
PCB-180	61.4 (17.8)	34.7–103	37.4 (0.64)	36.6–37.8	107 (27.7)	90.0–183	55.9 (10.3)	58.4–64.6
PCB-194	9.57 (18.1)	nd–63.1	5.31 (9.19)	nd–15.9	19.2 (35.0)	nd–87.3	8.66 (15.0)	nd–26.0
∑PCBs	249 (56.7)	164.3–336	109 (13.7)	95.3–123	467 (116)	282–642	198 (10.3)	189–209
∑(OCPs + PCBs)	357 (77.8)	257–493	130 (12.7)	117–142	667 (136)	468–869	252 (13.2)	237–262

nd: Not detected, and assumed as 0 for the calculation of total PCB and total OCP values.
nq: Not quantifiable.
SD: Standard deviation.

Table 3
Mean concentrations of OCPs and PCBs (ng g^{-1} lipid wt) in fish muscles collected from various countries.

Location	Species	HCB	HCHs	DDTs	PCBs (min–max)	References
Adriatic Sea, Italy	Red mullet	5.00	15.0	160	549 (369–863)	Stefanelli et al. (2004)
Shadegan Marshes, Iran	Abu mullet	2.60	145	155	130 (74–220)	Davodi et al. (2011)
Adriatic Sea, Italy	Red mullet	na	na	183	411	Perugini et al. (2004)
Gulf of Naples, Southern Italy	Grey mullet, sea bass	na	24.1	596	9340 (1042–31274)	Naso et al. (2005)
Marmara Sea, Turkey	Mullet	8.71	90.3	810	509	Coelhan et al. (2006)
Ebro Delta, Spain	Red mullet, sea mullet, sea bass	284	na	481	524 (109–800)	Pastor et al. (1996)
Atlantic coast, USA	Rock sea bass	0.12	na	9.56	41.5 (12–71)	Sajwan et al. (2008)
Ria de Aveiro, Portugal	Sea bass	na	na	108	275	Antunes and Gil (2004)
Tunis Bay, Tunisia	Golden grey mullet	na	na	na	(185–5566)	Masmoudi et al. (2007)
Bizerte Lagoon, Tunisia	Mullet and sea bass	8.37	12.1	134	358 (164–642)	Tunisia (this study)

na: Not available.

Quotients (TEQs) values of the analyzed congener were 0.08 and 0.12 pg (g w wt)$^{-1}$ respectively for mullet and sea bass from Bizerte Lagoon. In the Mediterranean Sea, TEQ values were 0.02 and 0.05 pg (g w wt)$^{-1}$ respectively for mullet and sea bass. There was a slight difference in PCB 118 TEQ values between the two fish species. Moreover the fish from Bizerte Lagoon have higher TEQ values compared to those from the Mediterranean Sea.

As specified in EC Regulation 1881/2006 (Official Journal of the European Union, 2006) the maximum permissible level for human consumption is of 8 pg (g w wt)$^{-1}$ of toxic equivalents (WH0-TEQ), for the sum of dioxins and dioxin-like PCBs in the muscle meat of fish and fishery products. The data here showed that the mean concentration of PCB 118 (0.1 and 0.04 pg TEQ (g w wt)$^{-1}$ respectively in the studied fish species from Bizerte Lagoon and the Mediterranean Sea) in all samples were below the legal limits. Further studies should assess other contaminants, such as other dioxin like PCB congeners, polychlorinated dibenzo-p-dioxins and polycholorinated dibenzofurans in Tunisian fish.

Concentrations of the target PCB congeners in Bizerte Lagoon fish are lower than those reported in fish from Mediterranean aquatic ecosystems such as Tunis Bay in Tunisia (Masmoudi et al., 2007) and Gulf of Naples in Italy (Naso et al., 2005). These levels are higher than those found in fish, from the Ria Aveiro in Portugal (Antunes and Gil, 2004), the Atlantic coast in USA (Sajwan et al., 2008) and from the Shadegan Marshes in Iran (Davodi et al., 2011). Comparable levels were reported in other fish specie from the Adriatic Sea in Italy (Perugini et al., 2004) (Table 3).

Although, these organochlorine compounds have been banned for the use and approval since 1980 and have been regulated in Tunisia in agreement with the Stockholm convention in 2001 (Stockholm Convention on Persistent Organic Pollutants, 2001), they are still detected in the Tunisian marine environment. This is due to their lipophilicity, stability, and persistence that facilitate their adsorption to sediments and their accumulation in the aquatic ecosystem.

3.5. Dietary intake of OCs from fish consumption

The estimated daily intake (EDI) of PCBs and OCPs through the studied fish species consumption for the general population in Bizerte (Northern of Tunisia) is summarized in Table 4. There was a slight difference in EDI values between the two studied fish species in both investigated areas. The fish from Bizerte Lagoon resulted in higher intakes of OCs compared to those from the Mediterranean Sea (Table 4). DDTs and PCBs have the highest contribution of all of OCs. DDTs accounted for 26.7% and 25.9% of the intake of all OCs respectively in mullet and sea bass from Bizerte Lagoon. Whereas, they accounted for 11.8% and 12.4% of the intake of all OCs respectively in mullet and sea bass from the Mediterranean Sea. PCBs accounted for 69.7% and 70% of the intake of all OCs respectively in mullet and sea bass from Bizerte Lagoon. Whereas they accounted for 83.3% and 78.6% of the intake of all OCs respectively in mullet and sea bass from the Mediterranean Sea.

As shown in Table 4, the EDI of DDTs and γ-HCH by the people were far below the acceptable daily intake (ADI) recommended by the Food and Agriculture Organization of the United Nations/World Health Organization (FAO/WHO) indicating this intake would not pose a health risk in Bizerte at the present.

The dietary intakes of HCB, HCHs, DDTs and PCBs from the studied Bizerte Lagoon fish species, with respective values of 6.31, 8.75, 101 and 267 ng d^{-1} were lower to those in fish from Indonesia and Sweden (Darnerud et al., 2006; Sudaryanto et al., 2007). However, the dietary intakes of OCPs in the present study were similar to those in Italy (Stefanelli et al., 2004).

3.6. OCPs and PCBs interspecies and intersites comparison

The highest content of ∑(OCPs + PCBs) was obtained in sea bass, and the lowest in mullet (Table 2). There were significant differences ($p < 0.05$) in the concentrations of ∑(OCPs + PCBs) in the investigated species. The accumulation of OCPs and PCBs in organisms is a very complex phenomenon, which involves several factors such as physico-chemical characteristics of different organochlorines, species, feeding habits, ages, genders, tissues and sizes of organisms, and trophic positon in the food chain. Variations in the metabolic capacity to detoxify may also have an effect on the magnitude and the pattern of accumulated organochlorines in the investigated species. These interspecies differences in this study are possibly related to their feeding habits and their trophic position in the food chain. The low concentrations of ∑(OCPs + PCBs) in mullet may be attributed to its feeding habits being omnivorous and consuming mostly phytoplankton, and to its low trophic position in the food chain. In contrast, the sea bass is a top predator (carnivorous fish) and has a high trophic position in the food chain. This result is in accordance with those obtained by Kong et al. (2005) and by Nie et al. (2005). In both species the total OCP and PCB levels were significantly higher in Bizerte Lagoon than in the reference area (the Mediterranean Sea) (Table 2; $p < 0.05$). The relatively high organochlorine levels in Bizerte Lagoon are attributable to the many sources of agricultural, municipal, and industrial contamination in Bizerte region. In particular, these chemicals mainly arrive in the studied ecosystem as a consequence of evaporation, atmospheric fallout, surface run-off, and waste water discharges from the intensively cultivated areas, the densely populated urban centers, the large industrial complexes, and the many waste dumps clustered along the coast.

3.7. Correlation of ΣPCBs, HCB, ΣHCHs and ΣDDTs with fish characteristics

Since it has been reported that persistent organic pollutant (POP) accumulation can be affected by biological factors, we have

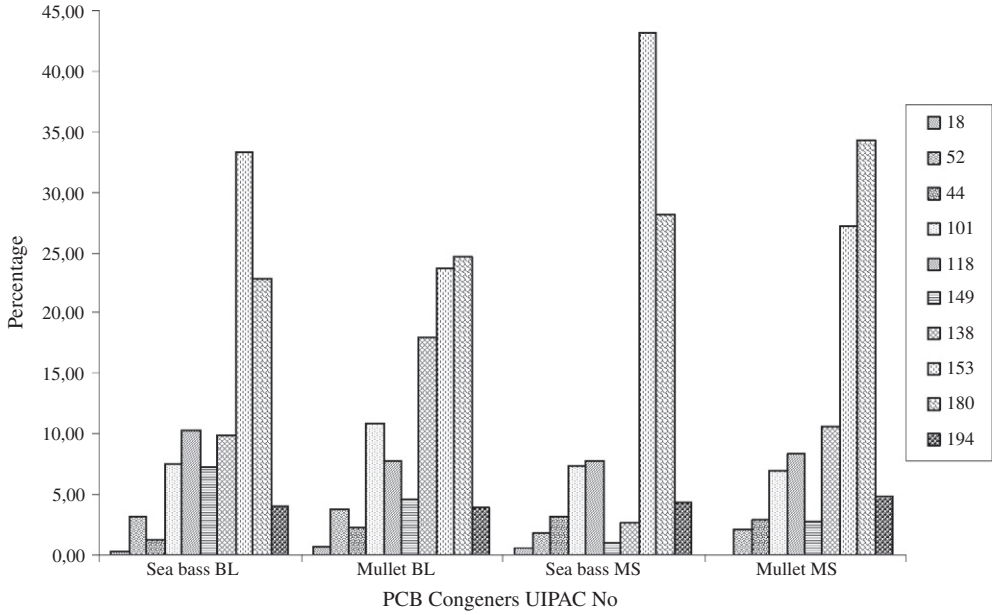

Fig. 2. Percentage contribution of individual polychlorinated biphenyls in muscle of Bizerte Lagoon (BL) and the Mediterranean Sea (MS) fishes.

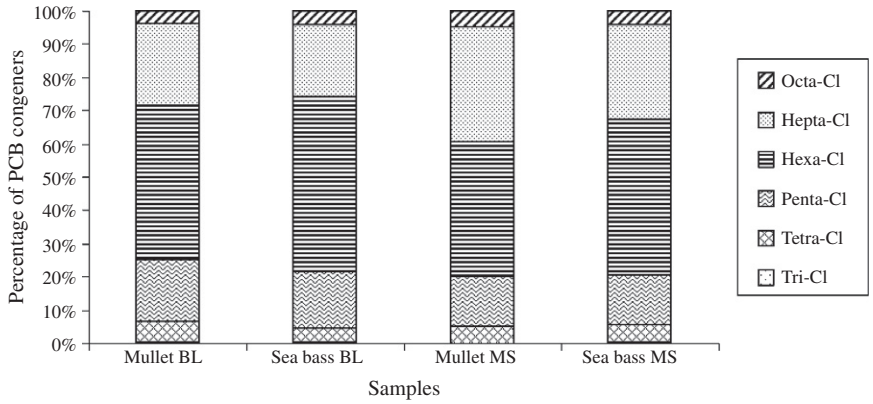

Fig. 3. The percentage of different Cl-substituted PCBs in the muscle of fishes from the Bizerte Lagoon (BL) and the Mediterranean Sea (MS).

Table 4
Estimated daily intakes of PCBs and OCPs through the studied fish species by human (average body wt 60 kg) in Bizerte (Northern Tunisia).

	EDI (ng (body wt)$^{-1}$ d^{-1})				ADI (ng (kg body wt)$^{-1}$ d^{-1}) (FAO/WHO)
	MC BL	DL BL	MC MS	DL MS	
HCB	0.10	0.11	0.04	0.18	
HCHs	0.11	0.18	0.02	0.15	8000 (γ-HCH)
DDTs	1.55	1.80	0.16	0.46	20000 (DDT)
PCBs	4.05	4.86	1.14	2.89	
∑OCs	5.80	6.95	1.35	3.68	

MC BL: *Mugil cephalus* from Bizerte Lagoon, MC MS *Mugil cephalus* from the Mediterranean Sea.
DL BL: *Dicentrarchus labrax* from Bizerte Lagoon, DL MS *Dicentrarchus labrax* from the Mediterranean Sea.
EDI: Estimated daily intake.
ADI: Acceptable daily intake.

correlated ∑PCBs, HCB, ∑HCHs and ∑DDTs concentrations with some specific characteristics of fish samples. As concerns PCBs, we revealed only a positive and statistically significant correlation between ∑PCBs concentrations and length and weight in both fish species. In mullet and sea bass, levels of ∑PCBs were correlated with length ($rs = 0.61$ and $rs = 0.72$ respectively $p < 0.05$) and with weight ($rs = 0.61$ and $rs = 0.83$ respectively $p < 0.05$). At the same time, a significant correlation between ∑PCBs concentrations and the lipid-content was observed only in sea bass ($rs = 0.67$ respectively $p < 0.05$). As concerns OCPs, we found only significant negative correlations between fish lipid content and HCB, as well as ∑HCHs levels. In mullet and sea bass, HCB was correlated with lipid content ($rs = -0.69$ and $rs = -0.59$ respectively $p < 0.05$). ∑HCHs was correlated with lipid content ($rs = -0.52$ and

rs = −0.59 respectively in mullet and sea bass $p < 0.05$). There was no correlation between fish lipid content, fish length, fish weight and ∑DDTs concentrations. These results were similar to those found by Deshpande et al. (2002), Chan et al. (1999) and by Ferrante et al. (2010). Note that the inverse relationship between fat content and pollutant levels is consistent with the results of other authors (Larsson et al., 1991; Ferrante et al., 2010) and provides support to the hypothesis that the pollutants are diluted in the increasing fat amounts of fishes.

4. Conclusions

OCPs and PCBs were detected in mullet and sea bass from Bizerte Lagoon. This is the first report of these compounds in marine fishes from Bizerte Lagoon. ∑OCPs and ∑PCBs in sea bass were higher than those in mullet in both investigated areas, a pattern which may be related to the different feeding habits and trophic position in the food chain of the two species. The organochlorine concentrations in the studied fish species from the Bizerte Lagoon were similar to or slightly higher than those reported for other species from other locations around the world. Significant negative correlations were observed between concentrations of sum HCHs and lipid content in fish species as well as between HCB concentrations and lipid content. Significant positive correlations were observed between concentrations of ∑PCBs and fish length as well as between ∑PCBs concentrations and fish weight. A significant correlation between ∑PCBs concentrations and fish lipid content was observed only in sea bass. The estimated daily intake for DDTs and γ-HCH via the studied fish species by the people of Bizerte were far below the ADI recommended by FAO/WHO, suggesting no relevant health risk by consuming fish.

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