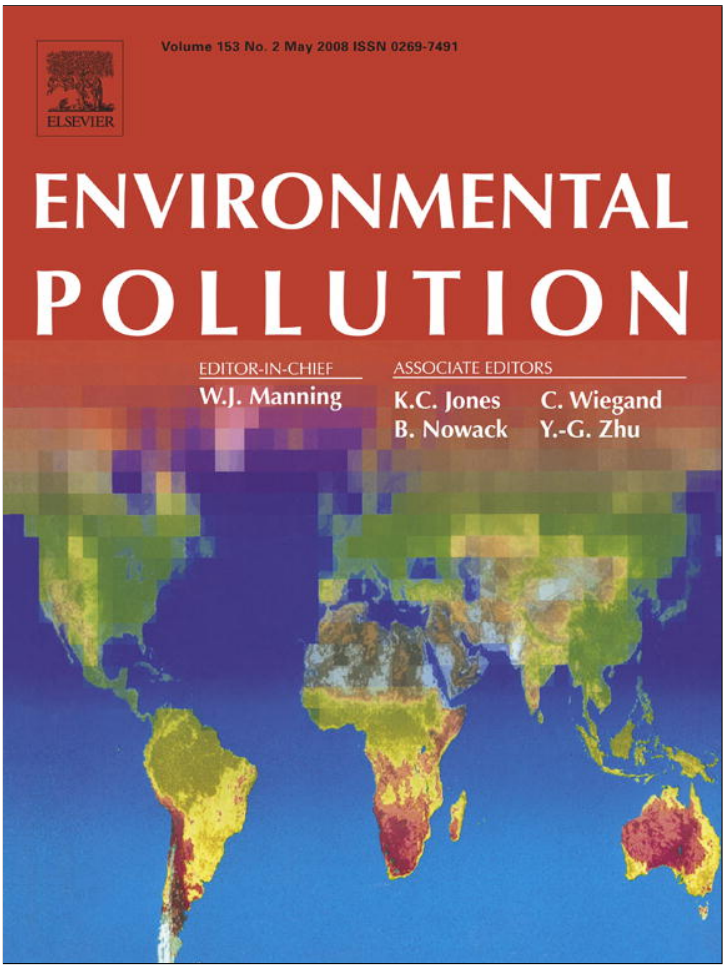

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Bioaccumulation of polychlorinated biphenyls in the eel (*Anguilla anguilla*) at the Camargue Nature Reserve – France

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The reserve of Camargue – South of France is impacted by a myriad of pollutant organic persistent like PCBs.

Abstract

Fish consumption is a potential source of human exposure to pollutants. Here, we study residue levels of PCBs in the eel, *Anguilla anguilla*, from the Nature Camargue Reserve in southern France. Chromatographic analysis (GC-ECD) found seventy identifiable congeners, among which, 10 are considered as dioxin-like PCBs, such as the non-ortho PCB 81 and the mono-ortho chlorobiphenyls PCB105, 114, 118, 123, 156, 157, 167, 170, 180. Toxic Equivalents (TEQ, WHO 2005 TEF-Toxic Equivalent Factors) varied among sites with a maximum in eels from Mornès (29.6 pg g^{-1} dry weight). Indicator PCBs (28, 52, 101, 118, 138, 153 and 180) were 22% and 29% of the total PCBs in livers and muscles respectively. Greater homogeneous bioaccumulation in muscle than that in liver suggests an increase risk for humans due to fish consumption.

Keywords: PCBs; Bioaccumulation; *Anguilla anguilla*; Camargue Reserve; Risk assessments

1. Introduction

Polychlorinated Biphenyls (PCBs) are contaminants released to environment due to antropogenic activities and have been reported as members of the group of ubiquitous, persistent, lipophilic, bioaccumulative and toxic or highly toxic microcontaminants (Falandysz et al., 2004). According to Baars et al. (2004) the waste disposal, both of house-holds and industrial wastes, is the major source of PCB emissions into the environment (Baars et al., 2004).

Fish consumption is a potential route of exposure for environmentally persistent organochlorine contaminants and the dietary uptake has contributed significantly to increase the human health risks (Turyk et al., 2006). In addition the traditional diet of human population in Europe plays an important role in the exposition to a PCB contamination (Johansen et al., 2004), where more than 90% of the total daily intake is derived from food (Papadopoulos et al., 2004). Recently the investigation in aquatic environment has revealed an increase in the bioaccumulation levels of PCBs in fish from different regions of the world (Falandysz et al., 2004; Ashizawa et al., 2005; Borga and Di Gardo, 2005; Evenset et al., 2005; Nakata et al., 2005; Stow et al., 2005; Vives et al., 2005; Ueno et al., 2005; Covaci et al., 2006).

The polychlorinated biphenyls can be accumulated in aquatic organism through the food chain, and its dietary intake by human population is already an evidence as demonstrated in studies with rats (Lehmler et al., 2005), human breast milk (Kunisue et al., 2006), adipose tissue of women (Vaclavik et al., 2006) and human serum levels (Hagmar et al., 2006).

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According to Priha et al. (2005) PCBs have mainly long-term effects and are expected to increase cancer risks, neurodevelopmental and hormonal effects in humans, also including immunosuppresion disturbs (Tryphonas et al., 1991). Our previous results (Oliveira Ribeiro et al., 2005) showed a high occurrence of chronic lesions as tumors in liver and spleen and high incidence of lipidosis in liver of *A. anguilla*. Other reported the effects of PCBs as endocrine hormone disrupter (Lin et al., 2006), oxidative stress agent (Murugesan et al., 2005), neurotoxic pollutant (Lehmler et al., 2005), cytochrome P4501A inductor (Behrens and Segner, 2005), a potent birth defects risk (Mendola et al., 2005) and immunomodulator (Iwanowicz et al., 2005).

Due to the complex nature of the 209 congeners the scientific community has been led to classified the representative congeners in two groups: the 'indicator PCBs' and the 'dioxin-like PCBs'. The 7 'indicator' congeners (IUPAC nos 28, 52, 101, 118, 138, 153, 180) have been chosen on the basis of their persistence in food web and their tendency to biomagnification. The 12 dioxin-like PCBs present a common mechanism of action with the polychlorinated dibenzo-*p*-dioxins (PCDDs) and the dibenzofurans (PCDFs): the bind ability to the aryl hydrocarbon (Ah) receptor. Consequently, it is now admitted that the World Health Organization Toxic Equivalency Factors (WHO-TEFs) are the best approach for risk assessment of halogenated aromatic hydrocarbons with dioxin-like properties (Van den Berg et al., 1998, 2005).

The present study will focus the residues levels of PCBs in *A. anguilla* from the Nature Reserve of Camargue – South of France, in order to increase the data about the potential human health risks associated with these contaminants due to fish consumption. This work is part of a large monitoring program supported by INRS and Camargue Nature Reserve and reports the results conducted from October 2003.

2. Materials and methods

2.1. Studied area and biological material

The Camargue Biosphere Reserve located in the Rhône delta – south of France, occupies 13,000 ha between the northern Vaccarès lagoon and the last undamaged sand dunes of the Mediterranean coast, is the largest coastal wetland area in Western Europe. The studied species was the European eel (*Anguilla anguilla*), a common predator fish living both in contact with sediments and in free waters. Thirty eels (weight 372 ± 39 g: length 55 ± 2 cm) were collected from three riparian locations: Fumemorte canal (FUM), 'La Capelière' (CAP) and the Mornès peninsula (MOR), the reference site (Fig. 1) in autumn (October 2003), and organs were carefully excised to chemical analyses.

2.2. Polychlorinated biphenyls extraction, separation, analyses procedures and characteristics

Livers and muscles were dissected for PCB congeners analysis and lipid fractions were extracted with a dichloromethane-methanol solution (Folch et al., 1957 modified method). Purification procedures were performed by means of solid phase extraction (SPE) on Florisil column according to the EPA method 3620 (Bond Elut Florisil, 1 g, 200 μM particle size – Varian) and analyses were performed by gas chromatography with AutoSystem XL (Perkin-Elmer), using electron capture detection (ECD 63Ni Source) and nitrogen as the carrier gas following an adapted procedure of the EPA Method 8081a. The detection limit ranged from 0.05 to 0.20 g kg^{-1} in fish tissues.

Among the 209 congeners, 7 compounds are considered as indicators PCBs (PCB 28, 52, 101, 118, 138, 153, 180), representing the major part of the PCB in biological tissues, and 12 compounds as dioxin-like PCB (non-ortho or the mono-ortho PCBs 77, 81, 105, 114, 118, 123, 126, 156, 157, 167, 169, 189 to which are added the PCBs 170 and 180).

2.3. TEQ evaluation

The TEQ of dioxin-like PCBs was calculated according Van den Berg et al. (1998) using the WHO-TEFs (World Health Organization – Toxic Equivalent Factors) for fish. The concept of TEF is based the following criteria: (1) show a structural relationship to the PCDDs and PCDFs; (2) bind to the Ah receptor (AhR); (3) elicit AhR mediated biochemical and toxic responses; and (4) be

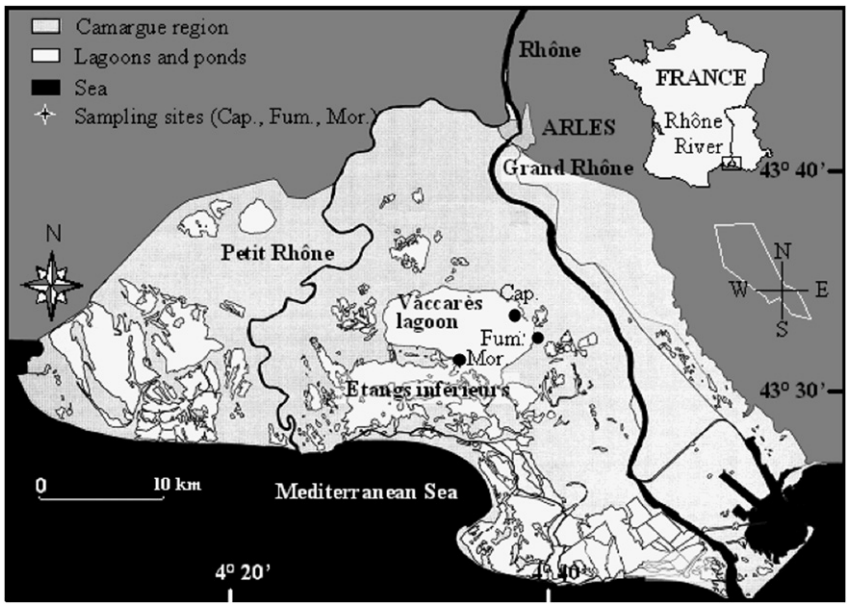

Fig. 1. The Biophere Camargue National Reserve – South of France: Mor – Mornès site; Cap – La Capellière site; Fum – Fumemorte site.

persistent and accumulate in the food chain. The evaluation of the WHO-TEFs for fish, quite different than those for mammals and birds, give the upper limit value for some PCB (<0.00005) (see Table 1), then we have calculated TEFs in excess (TEF = 0.00005).

2.4. Statistical procedures

Statistical analyses of PCBs data were achieved with the Statview program (version 5). Due to their non-Gaussian distribution, the data are presented as mean and extreme values. Intersite differences were checked using a non-parametric test, the rank-sum Mann–Whitney *U*-test. All tests were regarded as statistically significant when $p < 0.05$.

3. Results

The distribution of indicator and dioxin-like PCBs in eel muscles and livers from the Camargue Reserve is summarized in Fig. 2 and Table 1. Analysis of lipophilic pollutants in eels from the Vaccarès Lagoon demonstrates clearly inter-site variations of the PCB contamination (Fig. 2, Table 1). Concentration was greater in eels from Mornès and La Capelière than from Fumemorte (Fig. 2). Inter-tissue variation regardless of PCB type is evident in bioaccumulation profiles (indicator, dioxin-like or otherwise), especially in the congeners 19, 52/69, 123, 101/150, 138. Congeners 153 (muscle) and 138 (liver) reached levels >100 ng g^{-1} dry weight (all values are in dry weight). As in muscle, congeners 19 and 138 were most found in livers (>90 ng g^{-1}, Table 1).

PCBs in eels from La Capelière were similar in muscles and livers (1300 and 1600 ng g^{-1}, respectively), in contrast with both Mornès and Fumemorte, where bioaccumulation in muscles is greater than in livers (2700 vs. 1300 ng g^{-1}, in eels from Mornès; 1600 vs. 900 ng g^{-1}, in eels from Fumemorte, both $p < 0.05$, Table 2). Also, PCB bioaccumulation is more homogeneously distribution in livers than muscles. Individual variability in PCB bioaccumulation is more evident in liver than muscle. These differences seem to be due to indicator PCBs. Indeed, seven indicator congeners were found, often in high concentrations (in order of decreasing concentrations: 52 > 153 > 138 > 123 > 101 > 180/118, Table 1). Indicator PCBs comprised from 4.2% (in liver at Fumemorte) to 45% (muscle from La Capelière) of the total PCBs, with averages of 22% and 29% in livers and muscles respectively, within the entire eel population. Fumemorte typically had eels with lower concentrations (vs. La Capelière and Mornès, both $p < 0.05$; Table 2).

Lower concentrations of ∑di-CB, ∑hepta-CB and ∑octa-CB were found in both tissues from all studied sites, with little or no bioaccumulation, while the congeners ∑tri-CB, ∑tetra-CB, ∑penta-CB, and ∑hexa-CB were more concentrated (Fig. 3). Bioaccumulation of PCBs was less in eels from Fumemorte than Mornès (liver) or La Capelière (muscle).

Dioxin-like PCBs comprised a small part of total PCBs, from 0.13% in muscle in one individual from Fumemorte and 17% in fish liver from La Capelière, with an average of ~8%. These dioxin-like congeners were the PCBs 81, 105, 114, 118, 123, 156, 157, 167, 170, 167 and 180. The congeners contributing most to the WHO-TEQ, the PCB 126 and 169 were not detected. Individuals from Mornès had greater concentration of total PCBs in muscle than those from La Capelière ($p < 0.05$). This variation is notably due to the indicator PCBs and the dioxin-like PCBs with greater bioaccumulation levels in muscle at Mornès (Table 2).

WHO-TEQs (1997) values were rather low (Table 2). The greatest mean values were from La Capelière (liver) and Mornès (muscle; 4.4 and 10 pg g^{-1} dry weight, respectively), while the greatest level was from La Capelière (24 pg g^{-1} dry weight). The recent revised amount of admissible WHO toxic equivalents in foodstuffs is 4.0 pg g^{-1} fresh weight in the eel muscle, i.e. about 12.3 ± 0.5 pg g^{-1} dry weight, since muscle contains 67.5 ± 1.1 per cent water. In our study, only two eels from each, Mornès and La Capelière had TEQ levels above that limit (Van den Berg et al., 2005, Table 1).

4. Discussion

Polychlorinated biphenyls (PCBs) are aromatic synthetic chemicals, yet they have become common in the biosphere of many parts of the world. While no evidence yet links human death to acute exposure to PCBs, long term occupational exposure can increase mortality from cardiovascular disease and cancer (Pavuk et al., 2004). PCBs are lipophilic compounds, with a K_{ow} from 4.69–8.05 following their chlorination level (Zhou et al., 2005). Thus, accumulation patterns of each congener is expected to vary since their chemical and physical properties vary. In general, fish and shellfish consumption has contributed significantly to human exposure to these compounds, but more information is necessary to better understand patterns of effects due to different PCB congeners. Therefore, wildlife may be regarded as indicators for human health, providing evidence useful in a human health risk assessment and in identifying new sources of potential cancer-causing agents.

The potential effects of PCBs to human and wildlife is suggested by the serum reaction to adipose tissues that is reported for various PCB congeners (74, 99, 118, 138, 146, 153, 156, 167, 170, 180, 183 and 187). This suggests that humans, aquatic mammals, birds, fish and other biota have a similar PCB profile (ATSDR, 2000). Here, all congeners cited, except CB99, were found in *A. anguilla* from Camargue Reserve, and so fish consumption can be an important source of human exposure to PCBs. From 12 to 14 PCBs are regarded as being particularly toxic because of their ability to bind the aryl hydrocarbon receptor (AhR), a cytosolic receptor protein in most vertebrate tissues. Notably, AhR has a high affinity for 2, 3, 7, 8-substituted PCDD/Fs and some non-ortho and mono-ortho-substituted PCBs. They are called dioxin-like PCBs ranked as a function of the Toxic Equivalent Dioxin (TEQ, Van den Berg et al., 1998, 2005) and include 126, 169, 114, 156, 157, 77, 81, 105, 118, 123, 189, 167 plus 170 and 180. Neither PCB126 nor 169 were found in this study where the majority of eels had low TEQs. Still, risk evaluation based on TEQ dioxin is still debated since some (e.g., PCB126) have low response levels for enzyme induction in human cells, including primary hepatocytes, breast cancer cell lines and primary lymphocytes (Zeiger et al., 2001).

Table 1
Summary of PCBs compounds found in liver and muscle of *Anguilla anguilla* collected in Camargue Reserve − South of France, at October, 2003

Tissue site	1997 WHO-TEFs for fish*	Liver				Muscle		
		La Capelière (8)	Fumemorte (10)	Mornès (10)		La Capelière (11)	Fumemorte (9)	Mornès (8)
CB8		15.0 [1.1−63.4]	14.6 [2.5−31.3]	20.3 [9.2−59.6]	CB8	14.7 [3.3−43.7]	27.9 [2.2−65.5]	18.8 [6.9−46.9]
CB14		20.8 [2.3−75.2]	75.4 [nd−387]	26.5 [nd−127]	CB14	8.2 [nd−48.0]	9.2 [3.3−26.5]	8.9 [0.0−20.2]
CB13/15		11.4 [nd−21.0]	71.4 [8.6−306]	29.3 [2.2−81.4]	CB13/15[a,b]	4.3 [0.2−16.3]	28.9 [15.5−65.3]	27.6 [2.4−69.1]
CB19[b,c]		56.8 [20.7−94.5]	81.1 [21.2−133]	27.1 [nd−70.4]	CB19[a,b]	48.4 [0.8−128]	130.4 [nd−240]	252.2 [11.7−596]
CB18		9.2 [nd−24.1]	4.4 [nd−13.4]	6.0 [nd−15.8]	CB18[a]	2.8 [nd−10.4]	32.3 [1.9−101.7]	10.0 [nd−48.4]
CB17		11.9 [nd−27.0]	26.0 [nd−92.0]	15.9 [nd−49.7]	CB17	7.8 [nd−22.7]	18.7 [nd−48.8]	15.4 [nd−42.0]
CB32		3.0 [nd−6.1]	2.1 [nd−11.6]	2.5 [nd−6.2]	CB32[a,b]	2.5 [nd−5.9]	7.8 [4.2−19.4]	12.9 [0.6−30.6]
CB16		20.8 [nd−115]	3.2 [nd−9.1]	16.6 [nd−60.1]	CB16[a]	7.7 [nd−35.3]	8.3 [2.6−20.3]	6.5 [nd−19.2]
CB26/50[a,c]		20.5 [2.2−54.2]	103.0 [11.3−311]	30.3 [nd−99.9]	CB26/50[b]	20.4 [nd−51.8]	37.7 [9.8−87.5]	81.0 [13.9−141]
CB25		60.7 [2.4−184.8]	60.6 [3.7−213.0]	86.1 [3.6−410]	CB25	14.6 [nd−71.4]	14.0 [3.1−36.8]	13.2 [1.8−33.3]
CB51		5.3 [nd−17.9]	5.5 [nd−35.6]	5.5 [nd−34.2]	CB51	8.9 [nd−27.7]	3.8 [nd−19.8]	1.7 [nd−10.1]
CB31[c]		24.7 [nd−173]	0.8 [nd−8.2]	9.0 [nd−32.2]	CB31	13.7 [nd−30.7]	14.2 [3.6−65.8]	10.4 [2.0−27.5]
CB28[a]		*76.2 [4.9−274]*	*13.1 [1.9−37.2]*	*17.3 [nd−46.9]*	*CB28*	*63.0 [nd−202]*	*68.8 [19.3−296]*	*36.7 [6.1−149]*
CB36		17.6 [nd−59.7]	9.3 [1.8−22.4]	26.6 [nd−98.6]	CB36	10.0 [2.1−22.8]	9.6 [4.0−21.0]	16.4 [2.0−34.2]
CB20/33/45[c]		9.2 [nd−23.5]	4.8 [1.6−10.9]	10.4 [0.9−19.1]	CB20/33/45	7.8 [0.3−19.5]	11.9 [3.8−49.8]	5.9 [1.2−12.1]
CB22[c]		9.8 [nd−34.3]	22.2 [5.2−71.4]	17.0 [0.7−71.2]	CB22[a]	12.0 [3.3−33.4]	2.6 [nd−7.1]	13.5 [5.9−26.9]
CB52/69[c]		*25.0 [12.4−60.6]*	*35.7 [6.4−214]*	*34.6 [17.0−60.6]*	*CB52/69*	*83.1 [13.9−443]*	*37.8 [21.6−116]*	*229.1 [13.8−843]*
CB43/49		11.3 [2.2−17.7]	7.5 [nd−18.8]	14.0 [2.7−30.7]	CB43/49	12.7 [1.7−27.4]	10.0 [3.9−22.2]	5.6 [nd−15.0]
CB47/48/65[c]		4.0 [nd−10.5]	1.5 [nd−3.5]	6.1 [1.4−13.7]	CB47/48/65[c]	9.1 [nd−37.6]	7.5 [2.7−22.8]	2.4 [nd−8.4]
CB44[a,c]		8.8 [0.6−28.6]	2.0 [nd−5.3]	6.3 [1.2−12.7]	CB44	10.5 [nd−36.5]	34.6 [4.0−201]	12.1 [nd−18.5]
CB42		5.2 [nd−24.8]	2.7 [nd−6.6]	3.1 [nd−14.1]	CB42	4.0 [nd−17.5]	10.6 [0.5−34.8]	4.3 [0.14−10.0]
CB64/72		8.5 [2.0−17.8]	8.3 [1.4−20.7]	13.3 [1.1−31.6]	CB64/72[a]	9.1 [nd−28.9]	17.4 [11.1−27.0]	12.7 [4.5−20.0]
CB71/103		5.9 [nd−22.4]	3.3 [nd−9.3]	7.9 [nd−18.8]	CB71/103[c]	22.9 [nd−92.3]	13.6 [1.8−53.8]	4.3 [nd−19.7]
CB37/41/68		13.5 [nd−99.6]	0.9 [nd−4.1]	0.4 [nd−1.2]	CB37/41/68	3.1 [nd−18.3]	0.6 [nd−1.9]	1.7 [nd−5.9]
CB40		0.5 [nd−2.6]	0.2 [nd−2.1]	0.4 [nd−2.2]	CB40[a,c]	0.02 [nd−0.15]	2.0 [nd−6.7]	nd
CB95		45.1 [12.8−122]	18.8 [nd−43.7]	68.1 [nd−382]	CB95[b,c]	5.5 [nd−17.8]	1.7 [nd−5.9]	27.0 [1.0−92.8]
CB74		59.1 [6.1−218]	33.6 [nd−64.3]	38.1 [4.5−83.1]	CB74	37.8 [2.9−89.2]	58.6 [16.9−168]	60.5 [nd−138]
CB70[c]		61.4 [12.3−18]	20.4 [7.3−34.9]	45.5 [4.7−84.4]	CB70	52.8 [2.2−139.3]	62.4 [8.0−180]	47.2 [12.3−119]
CB76/80[c]		70.1 [20.4−184]	15.7 [nd−54.2]	60.6 [4.1−136]	CB76/80[a]	52.4 [4.8−129]	64.9 [nd−211]	90.8 [16.0−199]
CB66[c]		5.9 [nd−17.7]	7.7 [nd−41.7]	2.5 [nd−14.9]	CB66	12.9 [nd−90.5]	7.6 [nd−21.7]	16.0 [nd−53.9]
CB92[c]		35.9 [nd−128]	10.8 [nd−25.6]	27.5 [nd−55.0]	CB92	28.9 [1.4−74.5]	37.4 [nd−140]	48.5 [6.2−85.2]
CB84		4.1 [nd−16.3]	1.8 [nd−9.4]	3.0 [nd−16.3]	CB84	2.9 [nd−6.8]	10.8 [nd−46.5]	9.5 [nd−22.6]
CB101/150[a,c]		*44.5 [2.1−143]*	*6.7 [nd−19.5]*	*35.5 [3.8−100]*	*CB101/150*	*43.5 [5.3−89.1]*	*51.6 [4.5−120]*	*127.0 [12.1−278]*
CB60/113		12.5 [3.3−41.0]	7.3 [nd−14.7]	13.7 [1.9−26.9]	CB60/113	9.2 [0.8−22.9]	24.4 [nd−86.1]	23.5 [2.9−45.0]
CB112/119		14.7 [nd−109.3]	0.5 [nd−3.5]	2.8 [nd−7.9]	CB112/119	1.1 [nd−5.3]	0.2 [nd−1.2]	3.6 [nd−12.2]
CB148		7.3 [nd−36.7]	1.0 [nd−2.9]	1.6 [nd−7.8]	CB148	3.3 [nd−15.5]	0.7 [nd−3.4]	2.5 [0.1−8.4]
CB97/117[a,c]		9.6 [nd−27.2]	1.8 [nd−4.4]	7.2 [1.1−16.1]	CB97/117	10.2 [1.2−48.0]	6.5 [nd−16.4]	37.8 [1.6−101.4]
CB87/115[a,c]		11 n,0.1 [1.1−42.6]	2.3 [nd−4.5]	9.8 [0.8−26.2]	CB87/115[b]	11.3 [1.0−24.4]	14.9 [nd−43.0]	32.6 [3.2−60.5]
CB85		10.5 [nd−42.6]	3.1 [nd−13.0]	1.2 [nd−3.1]	CB85[b]	4.8 [nd−13.4]	15.8 [nd−46.9]	20.4 [0.3−72.9]
CB81[a,c]	**0.0005**	**7.2 [nd−24.7]**	**0.1 [nd−1.1]**	**2.8 [nd−15.8]**	**CB81[a,c]**	**12.7 [1.0−45.9]**	**4.2 [nd−22.6]**	**18.0 [2.4−36.1]**
CB110[a,c]		33.0 [7.9−90.4]	7.7 [2.6−20.5]	26.0 [3.1−60.4]	CB110[b]	28.5 [3.3−59.6]	27.3 [nd−57.8]	75.6 [9.3−139]
CB151[a,c]		24.7 [nd−94.2]	2.5 [nd−10.7]	24.2 [2.1−76.8]	CB151[b]	29.5 [3.1−66.2]	29.6 [nd−62.1]	69.5 [5.3−123]
CB135/144[a,c]		18.9 [nd−82.8]	2.7 [nd−12.6]	11.8 [1.7−55.7]	CB135/144	20.5 [2.2−46.2]	20.8 [nd−41.3]	53.9 [3.7−95.9]
CB123[a,c]	**< 0.000005**	**68.3 [14.8−226]**	**12.5 [1.6−50.3]**	**55.5 [4.6−159]**	**CB123[b]**	**46.0 [5.3−112]**	**58.5 [nd−119]**	**134 [10.2−231]**
CB118[b,c]		***48.2 [nd−100.7]***	***16.8 [nd−38.7]***	***15.4 [nd−147]***	***CB118[a]***	***1.6 [nd−6.2]***	***0.4 [nd−4.0]***	***0.5 [nd−3.7]***
CB146[b]		37.7 [nd−248]	11.4 [nd−63.7]	48.7 [nd−85.9]	CB146[c]	44.7 [nd−127]	76.6 [nd−253]	99.3 [6.1−257]
CB114[b]	**< 0.000005**	**4.4 [nd−12.3]**	**2.4 [nd−10.6]**	**4.6 [nd−14.6]**	**CB114[b]**	**0.6 [0.03−2.2]**	**6.0 [nd−24.3]**	**5.9 [0.2−17.2]**
CB132/179		13.4 [nd−78.4]	4.3 [nd−14.0]	13.0 [0.08−37.0]	CB132/179[c]	28.9 [1.8−75.8]	23.0 [nd−58.2]	41.3 [nd−92.7]
CB153[c]		*67.3 [0.3−370.0]*	*14.4 [1.2−70.9]*	*62.0 [2.5−254]*	*CB153[a,b,c]*	*69.2 [6.0−165]*	*14.0 [nd−73.2]*	*193.8 [22.1−369]*
CB141/176[b]		12.1 [nd−47.6]	2.6 [nd−23.4]	48.1 [nd−306]	CB141/176	13.9 [nd−47.1]	3.1 [nd−10.2]	23.1 [nd−96.2]

(*continued on next page*)

Table 1 (*continued*)

Tissue site	1997 WHO-TEFs for fish*	Liver				Muscle		
		La Capelière (8)	Fumemorte (10)	Mornès (10)		La Capelière (11)	Fumemorte (9)	Mornès (8)
CB105[c]	**< 0.000005**	**26.5 [1.4–107]**	**7.7 [nd–17.2]**	**21.3 [1.6–58.2]**	**CB105**	**26.9 [1.9–62.8]**	**37.7 [nd–111.1]**	**38.8 [7.4–119]**
CB137/141[b,c]		43.4 [nd–176]	7.2 [nd–26.7]	26.6 [3.5–112]	CB137/141[b]	30.8 [1.6–86.0]	51.2 [nd–96.2]	77.5 [5.3–136]
CB130/178		2.8 [nd–18.6]	0.1 [nd–1.5]	2.2 [nd–21.4]	CB130/178[c]	2.8 [nd–18.3]	0.1 [nd–0.7]	8.1 [nd–19.5]
CB163/164		2.7 [nd–8.6]	5.6 [nd–21.9]	2.9 [nd–16.8]	CB163/164[b,c]	1.8 [0.4–6.1]	0.5 [nd–1.9]	8.2 [1.1–25.1]
CB138[c]		*105 [nd–497]*	*21.0 [nd–110]*	*73.4 [7.3–273]*	*CB138*[c]	*101 [3.2–246]*	*64.2 [nd–169]*	*180.3 [22.9–392]*
CB175/182/187[b,c]		17.4 [nd–66.1]	2.5 [nd–21.9]	11.4 [nd–58.8]	CB175/182/187[b,c]	1.2 [nd–5.0]	13.6 [nd–114.8]	26.6 [1.2–55.0]
CB166/183[b,c]		6.8 [nd–36.5]	0.9 [nd–8.7]	3.0 [nd–22.0]	CB166/183[b,c]	0.5 [nd–2.1]	7.9 [nd–30.0]	20.4 [2.2–33.9]
CB159/185		0.5 [nd–2.8]	0.1 [nd–1.0]	0.1 [nd–0.4]	CB159/185	1.1 [nd–6.0]	15.9 [nd–69.8]	5.0 [nd–23.3]
CB128/162/174[c]		20.8 [nd–98.1]	5.1 [nd–32.0]	20.0 [1.0–60.9]	CB128/162/174	28.3 [1.5–131]	30.1 [nd–76.5]	56.0 [6.5–107]
CB177[c]		18.5 [nd–95.4]	2.8 [nd–21.6]	7.9 [0.7–35.6]	CB177[b]	8.4 [nd–25.6]	31.3 [nd–131]	59.2 [1.6–130]
CB197/201		5.4 [nd–13.5]	1.9 [nd–14.4]	2.2 [nd–10.9]	CB197/201[b,c]	8.6 [0.02–31.5]	0.7 [nd–1.6]	8.6 [0.7–20.0]
CB167[b]	**< 0.000005**	**0.7 [nd–3.6]**	**nd**	**0.3 [nd–3.2]**	**CB167**	**0.1 [nd–0.4]**	**0.1 [nd–1.2]**	**0.1 [nd–0.8]**
CB156/157	**< 0.000005**	**10.0 [nd–77.4]**	**0.6 [nd–5.8]**	**1.3 [nd–7.1]**	**CB156/157[b,c]**	**7.5 [nd–65.0]**	**0.7 [nd–1.6]**	**16.6 [0.2–62.6]**
CB180	**0.000005**	***26.5 [nd–146.2]***	***4.4 [nd–40.9]***	***10.5 [nd–45.4]***	***CB180***	***13.4 [0.8–53.8]***	***29.7 [nd–72.9]***	***48.5 [0.8–95.1]***
CB199[b]	–	18.1 [nd–90.8]	nd	3.5 [nd–30.7]	CB199[b,c]	3.4 [nd–20.5]	4.4 [nd–21.8]	25.0 [0.4–69.0]
CB170[b]		**5.7 [nd–19.7]**	**1.5 [nd–14.6]**	**1.1 [nd–5.4]**	**CB170**[c]	**7.9 [nd–34.7]**	**0.8 [nd–5.4]**	**5.9 [nd–16.5]**
CB190/203		2.8 [nd–9.9]	0.5 [nd–5.4]	2.5 [nd–8.6]	CB190/203[b]	0.5 [nd–2.1]	0.7 [nd–2.5]	4.1 [0.2–10.0]
CB195		8.3 [nd–58.7]	0.9 [nd–4.6]	6.4 [nd–59.7]	CB195	0.9 [nd–7.2]	0.6 [nd–1.5]	16.1 [nd–118]
CB194		51.7 [nd–266]	23.5 [nd–71.2]	21.3 [2.7–58.8]	CB194	30.3 [0.9–121.8]	82.1 [9.0–208]	53.9 [4.2–147]
CB205		0.1 [nd–1.2]	nd	1.3 [nd–13.2]	CB205	nd	nd	nd

Data are expressed as mean and extreme values [min–max]. Conclusions of the Mann–Whitney *U*-test: significant intersite variations $p < 0.05$. a: La Capelière vs Fumemorte; b: La Capelière vs Mornès; c: Fumemorte vs Mornès. Italic characters: Indicator PCBs; bold characters: dioxin-like PCBs. * after van den Berg et al. (1998) the value 1 is attributed to 2,3,7,8-TCDD (tetrachlorodibenzodioxin).

Several dioxin-like compounds on oestrogen metabolism in the malignant MCF 7 and non-tumorigenic MCF 10, a human mammary epithelial cell lines, have been studied (Van Duursen et al., 2003).

The majority of PCBs congeners in eels from the Camargue Nature Reserve are compounds that are still lacking information on toxic potential for humans and wildlife. The greatest PCB concentrations (liver from La Capelière, muscle from Mornès, 4.5 μg g^{-1} dry weight) are greater than both, the AEL (Adverse Effect Level) of 0.007 μg g^{-1} day^{-1} responsible for neurobehavioral alterations in infant monkeys, and the LOAEL (Lowest Observed Adverse Effect Level) of 0.005 μg g^{-1} day^{-1} producing immunological effects in adults (ATSDR, Agency for Toxic Substances and Disease Registry, 2000).

PCB contamination varies widely from site to site. Fish with lower concentrations (indicator and dioxin-like PCBs) were sampled near the Fumemorte catchment which receives Rhone River irrigation waters from nearby agriculture. In addition, hepatic and muscular TEQ are also lower than from La Capelière or Mornès. Indeed, fish captured at Mornès, which should not have agricultura or urban water input, had bioaccumulated more PCBs, some of which were dioxin-like (for example 81, 123, 114, 167, 156/157). This unexpected contamination may be due to atmospheric transfer (Banas et al., 2005), to water currents linked to a complex hydrological balance (Chauvelon, 1998), or to biomagnification as top-predators (Iannuzzi et al., 1996; Persic, 2004).

In addition to dioxin-like congeners, the seven indicator PCBs (138, 153 and 28, 52, 101, 118, 180) provide a contamination image. According to WHO (2000), the PCBs 138 and 153 are the major congeners found in all animal samples (as here, where PCB 153 was in both liver and muscle). These cause histological damage in organs including the liver and are neurological and neurochemical toxic agents (Sánchez-Alonso et al., 2003; Duffy and Zelikoff, 2006). A high incidence of necrosis in liver, along with other, organochlorine pesticide contaminants, can occur due to these congeners (Oliveira Ribeiro et al., 2005). Congener 28, which may also damage organs, was found in liver and muscle in this study, and its bioaccumulation is greatest in muscle, and so implies a human health risk. Also, liver concentrations were above the minimal risk level (0.005 μg g^{-1}). Although the congeners 118, 138, 153, 156, 170, 180 and 185 had relatively high levels in porpoise carp, bird, oligochaete, shrimp and human fat and milk, congener 74 found here in liver and muscle, was the most abundant in human fat and serum (ATSDR, 2000). PCB 153 in the majority of species is due to its slow rate of biotransformation and elimination (Dip et al., 2003; Hoekstra et al., 2003). Therefore, consumption of fish from Vaccarès lagoon is a potential pathway for human health risk.

Effects in liver of rat, mice and monkeys were observed after exposure to PCB 153 and others (Vezina et al., 2004). Exposure to mixtures of PCBs causes a diversity of effects, including hormonal, immunological, neurological and reproductive system damage. PCBs may be cause tumor activity and the environmental exposure has been associated with cancer of adipose tissues (Pavuk et al., 2004).

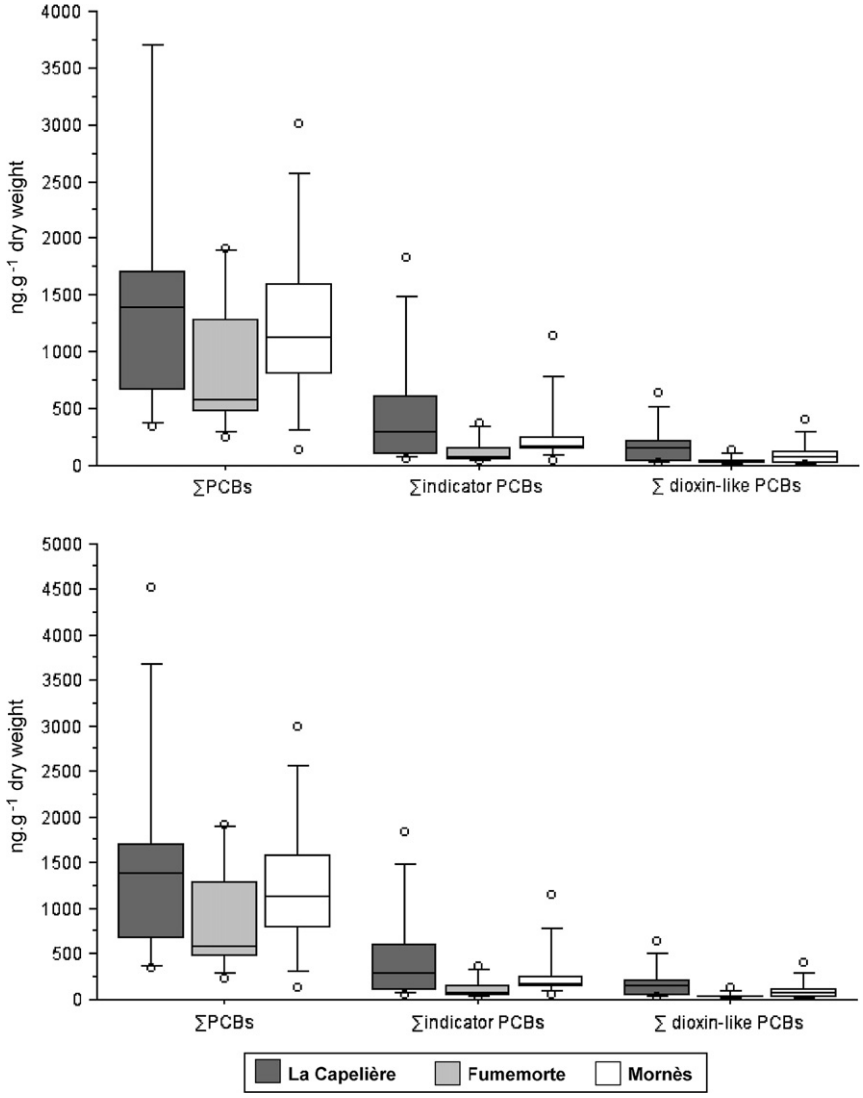

Fig. 2. Distribution of PCBs congeners in muscle and liver of *Anguilla anguilla* from Camargue Reserve – South of France, collected at October, 2003.

Wildlife response to exposure to PCBs and congeners varies widely, possibly reflecting not only variation in susceptibility but also different mechanisms of action or selective metabolism of individual congeners. PCBs tend to biomagnify in the food chain (Binelli and Provini, 2003; Burreau et al., 2004), reaching greater toxicity at higher trophic levels, such as in piscivorous and fatty fish (e.g., *A. anguilla*).

Table 2
Comparative total PCBs, indicator PCBs, dioxin-like PCBs and TEQ values in liver and muscle of *Anguilla anguilla* from Camargue Reserve – South of France collected at October 2003

	∑PCBs ng g^{-1} dw	∑indicator PCBs ng g^{-1} dw	% indicator PCBs	∑dioxin-like PCBs ng g^{-1} dw	% dioxin-like PCBs	TEQ pg g^{-1} dw
Liver		a,c*	a	a	a,b	a,c*
Vaccarès lagoon (28)	1499 [144–4539]	293 [43–1842]	22.1 [4.2–41]	110.1 [9–646]	8.3 [1.8–17]	2.02 [0.04–13]
La Capelière (8)	1556 [352–4539]	497 [68–1842]	28.1 [7–41]	190.3 [37–646]	11.6 [7–17]	4.4 [0.1–13]
Fumemorte (10)	855 [244–1912]	133 [43–375]	16.6 [4.2–30]	45.9 [9–139]	6.4 [1.8–11]	0.26 [0.04–1]
Mornès (10)	1281 [144–3005]	289 [51–1149]	22.7 [9–38]	110 [9–406]	7.6 [3.7–14]	1.89 [0.04–10]
Muscle	b	c*	a, c	b,c*		c
∑Vaccarès lagoon (27)	1803 [254–4477]	588 [54–1819]	29.3 [10–45]	158 [0.3–468]	8.5 [0.13–14]	6.5 [0.3–24]
La Capelière (10)	1290 [254–2680]	494 [54–1200]	32.9 [15–45]	107 [11–233]	8.6 [4.5–13]	7.1 [0.6–24]
Fumemorte (9)	1554 [774–2997]	331 [89–816]	21.0 [10–29]	134 [0.3–250]	7.9 [0.13–14]	2.6 [0.3–12]
Mornès (8)	2724 [515–4477]	996 [116–1819]	33.9 [20–41]	250 [23–468]	9.1 [4.9–13]	10 [1.3–20]

The data are expressed as mean and extreme values [min–max]. Conclusions of the Mann–Whitney *U*-test: significant intersite variations $p < 0.05$. a: La Capelière vs Fumemorte; b: La Capelière vs Mornès; c: Fumemorte vs Mornès. c* $p < 0.06$ Fumemorte vs Mornès.

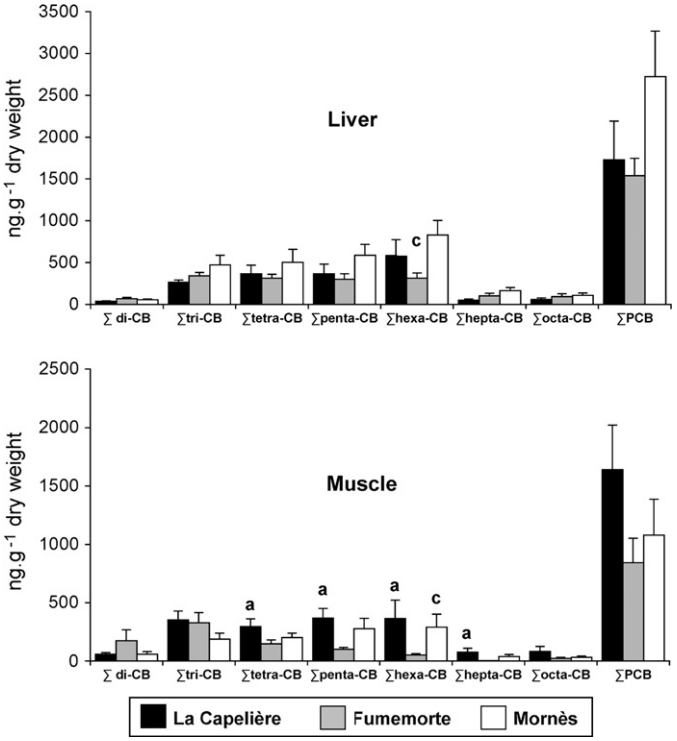

Fig. 3. Comparative distribution of di, tri, tetra, penta, hexa, hepta, octa groups and total PCBs in muscle and liver of *Anguilla anguilla* from La Capelère, Fumemorte and Mornès sites in Camargue Reserve – South of France, collected at October, 2003.

Sport fish consumption in the Great Lakes is therefore a potential source of exposure for environmentally persistent organochlorine contaminants that may cause human heath risks (Turyk et al., 2006). Associations occur between fish consumption and body burdens of PCBs within sports or professional fishermen, or others that consume large quantities of contaminated fish (Vaclavik et al., 2006). Advancements in TEQ calculations seem to provide an accurate evaluation of risk for PCBs and dioxin-like molecules in the environment (Alcock et al., 1998).

5. Conclusion

PCB exposure from fish consumption is an important source of human exposure and should be better studied. Since certain hydroxylated methylsulfonyl ($MeSO_2$) PCB metabolites may reach greater levels than their respective parent compounds, and therefore it is necessary to investigate potential biological or toxicological activities of these persistent metabolites. In addition, the persistence and mobility of these toxic chemicals indicate that the problems are global and that the solution must be at the global level.

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