

# POPs in *Sparus auratus* Species from the Eastern and Western Egyptian Mediterranean Coast: A Health Hazard Study

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## Abstract

The residual concentrations of HCHs, TC, DDTs, THC and PCBs were measured in *Sparus auratus* collected from the eastern and western sectors of the Egyptian Mediterranean Sea, as the most common fish species in the area. Gas chromatograph equipped with <sup>63</sup>Ni-electron capture detector and 35% phenyl polysilphenylenesiloxane capillary column was used for the analysis and quantification after well-established extraction techniques. The concentrations (wet weight) of PCBs, DDTs, TCs, HCHs were: (1.87 - 616.66 ng·g<sup>-1</sup>), (0.42 - 98.28 ng·g<sup>-1</sup>), (0.06 - 2.94 ng·g<sup>-1</sup>), (0.35 - 11.77 ng·g<sup>-1</sup>), respectively in *Sparus auratus* collected from the eastern sector. However, these concentrations as wet weight in *Sparus auratus* from the western sector were: (5.75 - 605.53 ng·g<sup>-1</sup>), (1.53 - 226.47 ng·g<sup>-1</sup>), (0.09 - 8.12 ng·g<sup>-1</sup>), (0.26 - 5.80 ng·g<sup>-1</sup>), respectively. In general, concentrations of pesticides and PCBs in *Sparus auratus* either from the western or eastern Egyptian Mediterranean coast were far below the international permissible levels.

## Keywords

Pesticides, PCBs, THC, *Sparus auratus*, Mediterranean, Egypt

## 1. Introduction

Contamination of the Egyptian Mediterranean Coast has become a subject of a great deal of research in recent years. The continuing and increasing release of a great number of industrial, agricultural, commercial and domestic waste effluents and emissions as well as hazardous substances have affected the characteris-

tics of water [1]-[7], sediments [8]-[15], and fish which is most commonly chosen because of the implications it carries for human consumption and health risk [16] [17] [18] [19] [20]. Major concern has been directed to persistent organic pollutants (POPs) as they are highly resistant to degradation by biological photolytic and/or chemical means [21] [22] [23]. These compounds were found widespread in the environmental media, humans and wildlife, and globally distributed including remote areas where they have never been used [24]. POPs are used to refer to organic compounds whose presence in the environment in very small amounts can cause significant harm to ecological systems and/or humans [25]. This study was aimed to compare the concentration levels of POPs in tissues of *Sparus auratus* as the most economical marine fish species in the eastern and western sectors of the Egyptian Mediterranean Sea.

## 2. Materials and Methods

Biota samples, *Sparus auratus* species, were collected fresh from 12 stations by fishermen working in the Eastern and western sectors of the Egyptian Mediterranean Sea during 2010 (Figure 1), no more than 2 h after catching and then kept frozen in the laboratory. Biological specimens were dissected and their tissues were kept frozen until extraction. Fish tissue (10 g of wet wt. of flesh) as placed in a blender, and 30 g of anhydrous sodium sulphate was added. Samples were blended at high speed until the mixture was well homogenized (2 - 3 min). The mixture was then transferred to a pre-cleaned extraction thimble and the dehydrated tissue was extracted with 200 ml (1:1) of n-hexane-dichloromethane for 8 h in a Soxhlet apparatus cycling 5 - 6 times h<sup>-1</sup>. Anhydrous sodium sulphate (30 g) was extracted in the same fashion as the sample and used as the blank. The extracted solvents were concentrated with a rotary evaporator and concentrated with a pure nitrogen gas stream down to a volume of 2 ml [26] [27]. Clean-up and fractionation were performed by passing the extract through a silica/alumina column. Elution was performed using 70 ml of hexane for PCBs congeners (F1), followed by elution with a 50 ml mixture containing 70% hexane and 30% dichloromethane for pesticide fraction (F2). Finally, eluted samples

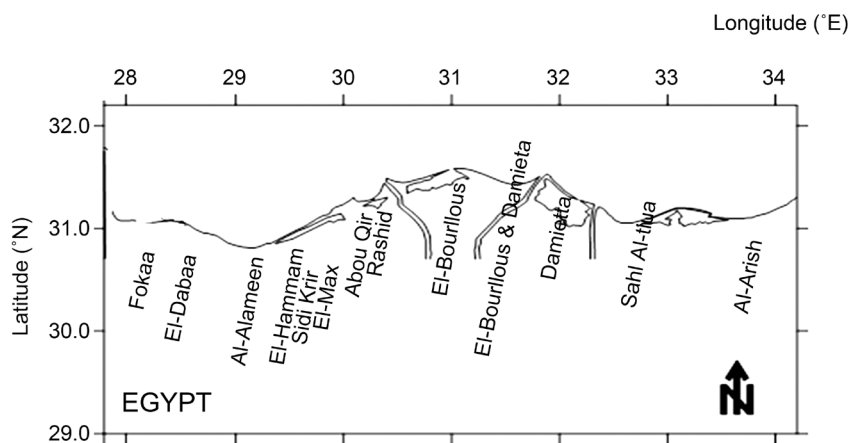


Figure 1. Sampling stations of the study area.

were concentrated under a gentle stream of purified nitrogen to about 0.2 ml, prior to injection into GC/ECD for analysis. All samples were analyzed by a Hewlett Packard 5890 series II GC gas chromatograph equipped with a  $^{63}\text{Ni}$  electron capture detector. The instrument was operated in split less mode (3  $\mu\text{L}$  split less injection) with the injection port maintained at 290°C and the detector maintained at 300°C. A fused- silica capillary column; Thermo TR-35 MS (30 m, 0.25 mm, 0.25  $\mu\text{m}$ ) with 35% phenyl polysilphenylenesiloxane was used for the quantification. The temperature was programmed from 90°C - 140°C with rate of 5°C min<sup>-1</sup>, then held at 140°C for 1 min, and from 140°C - 250°C with rate of 3°C min<sup>-1</sup> and was held at 250°C for 1 min, and from 250°C - 300°C with rate of 20°C min<sup>-1</sup> and was held at 300°C for 1 min. The injector and detector temperatures were set at 280°C and 310°C, respectively. Three  $\mu\text{L}$  volume of each sample was injected in the split less mode and the purge time was 1 min. To control the analytical reliability and assure recovery efficiency and accuracy of the results, four analyses were conducted on organochlorine compounds reference material SRM-2974 a freeze-dried muscle tissue (*Mytilus edulis*) provided by EIMP-IAEA. The laboratory results showed that recovery efficiency ranged from 90% to 105% with coefficients of variation of 8% - 15% for all organochlorine compounds. The limit of detection in the present study was estimated to be 0.014 ng.g<sup>-1</sup> for PCB and 0.014 ng.g<sup>-1</sup> for pesticides based on the minimum quantity of sample required for a discernible peak appeared on the chromatogram.

### 3. Results and Discussion

Muscles of fish showed the presence of a wide variety of organochlorines including  $\alpha$ ,  $\beta$  and  $\gamma$ -HCH, dieldrin, aldrin, endrin, DDT and their metabolites as well as PCBs (Table 1 and Table 2). The mean composition of OCPs in *Sparus auratus* was 17.96%, 6.46%, 75.54% for HCHs, TCs and DDTs, respectively. DDTs composition was much higher than HCHs and TCs, which might be attributed to the more bio-accumulative ability of DDTs (Table 1). Low concentrations of HCHs, which were recorded in fish tissues of the present study, reflect their lower potential for bioaccumulation than other pesticides. Furthermore, higher vapour pressures of HCHs facilitate relatively rapid atmospheric dissipation in the tropics, leaving fewer residues in soil and water [28]. The most striking difference in HCHs composition of the investigated *Sparus auratus* (approximately 18.30% of  $\alpha$ -HCH, 70.48% of  $\beta$ -HCH and 11.23% of  $\gamma$ -HCH) is the high level of  $\beta$ -HCH was the predominant isomer of HCHs which can exist in the environment for several years because they are stable, resistant to microbial degradation, and have a long half-life with a low solubility and vapour pressure [29]. Although, aldrin is readily converted to dieldrin in many environmental compartments, the mean composition of Aldrin, Dieldrin and Endrin in *Sparus auratus* of the present study was 13.87%, 19.08% and 67.05%, respectively. This indicates that there is a weathered source of Aldrin with sufficient oxygen available for the oxidation process to its epoxide form; Dieldrin. Endrin is highly

**Table 1.** Concentration of chlorinated pesticides, PCBs and THCs measured in *Sparus auratus* collected from the Eastern part of the Western Egyptian Mediterranean Coast during 2010.

Stations	Concentration, ng.g <sup>-1</sup>															
	$\alpha$ -HCH	$\beta$ -HCH	$\gamma$ -HCH	HCHs	Aldrin	Dieldrin	Endrin	TCs	$\alpha$ , p-DDE	p, p-DDE	$\alpha$ , p-DDD	p, p-DDD	$\alpha$ , p-DDT	p, p-DDT	DDTs	DDTs
Abou Qir	0.06	0.06	0.24	0.35	0.05	0.01	0.02	0.08	0.02	0.03	0.04	0.03	0.08	0.21	0.42	0.42
Rashid	0.21	1.85	2.15	4.21	0.83	0.05	1.96	2.83	ND	2.3	0.74	1.23	1.28	92.72	98.28	98.28
Bourlous	0.45	3.42	0.02	3.89	0.02	0.01	0.02	0.06	0.06	0.21	0.05	0.07	0.1	5.37	5.86	5.86
Bourlous & Damietta	0.83	4.09	0.03	4.96	0.12	0.03	2.29	2.44	0.01	0.17	0.02	1.32	0.33	1.27	3.13	3.13
Damietta	1.16	2.45	0.07	3.67	0.21	0.08	2.65	2.94	0.02	0.29	0.02	1.63	0.29	2.3	4.56	4.56
Sahl Altina	2.6	8.46	0.72	11.77	0.22	1.81	0.02	2.05	0.92	1.8	0.37	0.64	ND	5.44	9.16	9.16
AV	0.88	3.39	0.54	4.81	0.24	0.33	1.16	1.73	0.17	0.8	0.21	0.82	0.42	17.89	20.23	20.23
MAX	2.6	8.46	2.15	11.77	0.83	1.81	2.65	2.94	0.92	2.3	0.74	1.63	1.28	92.72	98.28	98.28
MIN	0.06	0.06	0.02	0.35	0.02	0.01	0.02	0.06	ND	0.03	0.02	0.03	0.08	0.21	0.42	0.42
Station	PCB 28	PCB 52	PCB 101	PCB 118	PCB 135	PCB 138	PCB 153	PCB 180	PCBs	TP	Total	THC				
Abou Qir	0.06	0.09	0.08	0.02	ND	0.43	0.79	0.4	1.87	0.85	2.72	410				
Rashid	2.72	0.37	0.11	3.06	ND	19.08	67.28	7.83	100.44	105.32	205.76	480				
El Bourlous	0.31	8.81	1.68	0.17	ND	0.95	8.57	0.38	20.88	9.81	30.69	370				
Bourlous & Damietta	0.38	67.81	0.27	0.04	ND	1.01	0.78	1.52	71.81	10.52	82.33	480				
Damietta	0.25	166.05	4.43	0.04	ND	1.15	0.62	0.25	172.79	11.17	183.97	590				
Sahl Altina	2.66	591.13	2.19	0.05	ND	7.8	12.35	0.49	616.66	22.98	639.64	350				
AV	1.06	139.04	1.46	0.56	ND	5.07	15.07	1.81	164.08	26.78	190.85	450				
MAX	2.72	591.13	4.43	3.06	ND	19.08	67.28	7.83	616.66	105.32	639.64	590				
MIN	0.06	0.09	0.08	0.02	ND	0.43	0.62	0.25	1.87	0.85	2.72	350				

AV = average, ND: <Detection Limit, HCHs =  $\alpha$ -HCH +  $\beta$ -HCH +  $\gamma$ -HCH, TCs = Aldrin + Dieldrin + endrin, DDTs =  $\alpha$ , p-DDE + p, p-DDE +  $\alpha$ , p-DDD + p, p-DDD +  $\alpha$ , p-DDT + p, p-DDT. TP: Total Pesticides. THCs = total petroleum hydrocarbons.

**Table 2.** Concentration of chlorinated pesticides, PCBs and THCs recorded in *Sparus auratus* collected from the Western part of the Eastern Egyptian Mediterranean Coast during 2010.

Stations	Concentration, ng.g <sup>-1</sup>														
	$\alpha$ -HCH	$\beta$ -HCH	$\gamma$ -HCH	HCHs	Aldrin	Dieldrin	Endrin	TCs	$\alpha$ , p-DDE	p, p-DDE	$\alpha$ , p-DDD	p, p-DDD	$\alpha$ , p-DDT	p, p-DDT	DDTs
Fokaa	0.17	0.07	0.02	0.26	0.02	0.02	0.05	0.09	0.01	0.31	0.04	0.05	0.27	0.85	1.53
El-Dabaa	0.21	0.32	0.01	0.53	2.16	0.06	5.9	8.12	0.01	19.26	8.22	6.73	17.29	108.92	160.41
Al-Alameen	0.05	0.57	0.01	0.64	1.46	0.06	1.92	3.44	0.02	20.8	10.82	5.47	35.05	154.31	226.47
El-Hammam	0.88	0.01	0.02	0.91	0.01	0.03	0.34	0.38	0.01	109.78	0.02	0.56	4.57	8.37	123.32
SidiKrir	0.73	3.36	0.01	4.1	0.3	0.01	2.46	2.77	0.02	1.97	0.02	0.06	4.21	2.36	8.64
El-Max	0.99	4.4	0.41	5.8	0.23	0.02	3.81	4.06	0.01	0.07	0.07	0.06	4.73	2.24	7.18
AV	0.5	1.45	0.08	2.04	0.7	0.03	2.41	3.14	0.01	25.36	3.2	2.16	11.02	46.17	87.92
MAX	0.99	4.4	0.41	5.8	2.16	0.06	5.9	8.12	0.02	109.78	10.82	6.73	35.05	154.31	226.47
MIN	0.05	0.01	0.01	0.26	0.01	0.01	0.05	0.09	0.01	0.07	0.02	0.05	0.27	0.85	1.53
Station	PCB 28	PCB 52	PCB 101	PCB 118	PCB 135	PCB 138	PCB 153	PCB 180	PCBs	TP	Total	THC			
Fokaa	0.12	0.11	0.05	0.04	ND	1.98	1.69	1.76	5.75	1.87	7.62	190			
El-Dabaa	0.02	0.6	0.03	21.41	ND	151.94	66.09	97.45	337.54	169.07	506.61	380			
Al-Alameen	0.04	0.76	0.09	35.23	ND	265.59	121.57	182.25	605.53	230.54	836.07	370			
El-Hammam	0.46	0.08	0.06	0.61	ND	10.07	11.73	5.39	28.4	124.61	153	390			
SidiKrir	0.1	391.79	4.84	0.92	ND	0.52	3.53	2.87	404.56	15.51	420.07	200			
El-Max	0.22	331.81	7.14	1.17	ND	0.75	4.26	7.5	352.84	17.04	369.89	430			
AV	0.16	120.86	2.04	9.9	ND	71.81	34.81	49.54	289.1	93.11	382.21	330			
MAX	0.46	391.79	7.14	35.23	ND	265.59	121.57	182.25	605.53	230.54	836.07	430			
MIN	0.02	0.08	0.03	0.04	ND	0.52	1.69	1.76	5.75	1.87	7.62	190			

AV = average, ND: <Detection Limit, HCHs =  $\alpha$ -HCH +  $\beta$ -HCH +  $\delta$ -HCH, TCs = Aldrin + Dieldrin + endrin, DDTs =  $\alpha$ , p-DDE + p, p-DDE +  $\alpha$ , p-DDD +  $\alpha$ , p-DDT + p, p-DDT. TP: Total Pesticides, THCs = total petroleum hydrocarbons.

toxic; it is much higher persistent than Dieldrin and its occurrence was expected to be widespread. Relative to HCHs, biota recorded high levels of DDTs showing that, p, p-DDT was the predominant isomer in fish species, with 88.43% of all DDTs. The mean compositions of o, p-DDE, p, p'-DDE, o, p-DDD, p, p'-DDD, o, p-DDT and p, p'-DDT in biota were 0.84%, 3.95%, 1.03%, 4.05%, 2.07% and 88.43%, respectively. DDD levels were higher than DDE suggesting that DDTs endure the anaerobic transformation in organisms. Metabolic transformation of DDT under anaerobic conditions, p, p'-DDD is formed [30]. The present study showed that the average concentration of DDTs recorded 75.54% of all OCPs concentrations. Several authors have observed preferential accumulation of DDT degradation products in aquatic organisms [31]. This phenomenon is due to uptake, metabolism and depuration mechanisms for DDT compounds in muscles [32]. The average ratio of HCHs to DDTs concentration in muscles was ~23%, suggesting excretion of HCHs in marine muscles. Accumulation of PCBs was more pronounced in tissues of *Sparus auratus* with 164.08 ng·g<sup>-1</sup>; wet weight. The average concentration of OCPs and PCBs in fish species showed a decreasing trend from PCBs > DDTs > HCHs > TCs. PCBs congeners 28, 52, 101, 118, 135, 138, 153 and 180 were 0.65%, 84.74%, 0.89%, 0.34%, ND%, 3.09%, 9.18% and 1.10%, respectively in fish species (Table 1). The results showed that these compounds probably originated via atmospheric deposition as stated by Doong *et al.* [33]. PCB52, PCB153, PCB138 and PCB180 were the most detected congeners in all investigated fish species. In contrast, PCB135 was not recorded in *Sparus auratus*. Contribution of low chlorinated congeners was minimal due to the fact that these congeners have an increased mobility from the substrate to water and therefore more available to aquatic organisms [34]. In addition, they are very susceptible to metabolism and are eliminated in marine environment. Thus, the order of decreasing concentrations in the eastern sector was: HCHs: Sahl Altina > El-Bourllous & Damietta > Rashid > El-Bourllous > Damietta > Abu-Qir. TCs: Damietta > Rashid > El-Bourllous & Damietta > Sahl Altina > Abu Qir > El-Bourllous. DDTs: Rashid > Sahl-Altina > El-Bourllous > Damietta > El- Bourllous & Damietta > Abou Qir. PCBs: Sahl-Altina > Damietta > Rashid > El- Bourllous & Damietta > El Bourlos > Abou Qir. High concentrations of THCs in 590 ng·g<sup>-1</sup> were measured in Damietta area. Concentrations of THCs in *Sparus auratus* varied in a narrow range from 350 - 590 ng·g<sup>-1</sup>; with an average of 450 ng·g<sup>-1</sup>.

On the same context, the mean composition of OCPs in *Sparus auratus* was 2.19%, 3.37%, 94.43% for HCHs, TCs and DDTs, respectively. DDTs composition was much higher than TCs and HCHs, which might be attributed to the more bio-accumulative ability of DDTs (Table 2). The most striking difference in HCHs composition of the investigated *Sparus auratus* (approximately 24.51% of  $\alpha$ -HCH, 71.08% of  $\beta$ -HCH and 3.92% of  $\gamma$ -HCH) is the high level of  $\beta$ -HCH was the predominant isomer of HCHs which can exist in the environment for several years as stated by Willett *et al.* [29]. Although, Aldrin is readily converted to Dieldrin in many environmental compartments, the mean composition of

Aldrin, Dieldrin and Endrin in *Sparus auratus* of the present study was 22.29%, 0.96% and 76.75%, respectively. This indicates that there is a fresh source of Aldrin with no sufficient oxygen available for the oxidation process to its epoxide form; Dieldrin. Endrin is highly toxic; it is much higher persistent than Dieldrin and its occurrence was expected to be widespread in fish species of the western sector. Relative to HCHs, biota recorded high levels of DDTs showing that, p, p-DDT was the predominant isomer in fish species, with 52.51% of all DDTs. The mean compositions of o, p-DDE, p, p'-DDE, o, p-DDD, p, p'-DDD, o, p-DDT and p, p'-DDT in *Sparus auratus* were 0.01%, 28.84%, 3.64%, 2.46%, 12.53% and 54.51%, respectively.

DDE levels were higher than DDD suggesting that DDTs endure the aerobic transformation in organisms. Metabolic transformation of DDT under aerobic conditions, p, p'-DDE is formed [30]. The present study showed that the average concentration of DDTs recorded 94.43% of all OCPs concentrations. The average ratios of HCHs to DDTs concentration in muscles was 2.32%, suggesting excretion of HCHs in marine muscles. Accumulation of PCBs was more pronounced in tissues of *Sparus auratus* with 289.10 ng·g<sup>-1</sup>; wet weight. The average concentration of OCPs and PCBs in fish species showed a decreasing trend from PCBs > DDTs > TCs > HCHs. PCBs congeners 28, 52, 101, 118, 135, 138, 153 and 180 were 0.06%, 41.81%, 0.71%, 3.42%, ND%, 24.84%, 12.04% and 17.14%, respectively in fish species (Table 2). The results showed that these compounds probably originated via atmospheric deposition as stated by Doong *et al.* [33]. PCB52, PCB153, PCB138 and PCB180 were the most detected congeners in all investigated fish species. In contrast, PCB135 was not recorded in *Sparus auratus* in the western sector as also happened in the eastern one. The order of decreasing concentrations in all *Sparus auratus* samples of the Western part was HCHs: El-Max > Sidi Krir > El-Hammam > Al-Alameen > El-Dabaa > Fokaa, TCs: El-Dabaa > El-Max > Al-Alameen > Sidi Krir. > El-Hammam > Fokaa, DDTs: Al-Alameen > El-Dabaa > El-Hammam > Sidi Krir > El-Max > Fokaa and PCBs: Al-Alameen > Sidi Krir > El-Max > El-Dabaa > El-Hammam > Fokaa. Maximum concentrations of THCs were observed at El-Max; 430 ng·g<sup>-1</sup>, El-Hammam; 390 ng·g<sup>-1</sup>, El-Dabaa; 380 ng·g<sup>-1</sup> and Al-Alameen; 370 ng·g<sup>-1</sup>. THCs varied from 190 - 430 ng·g<sup>-1</sup>, with an average of 330 ng·g<sup>-1</sup> (Table 2).

The maximum permissible levels of toxic DDTs, PCBs and TCs recommended by the National Academy of Sciences and National Academy of Engineering [35] for the protection of aquatic biota are 1000 - 5000 ng/g for PCBs and 100 ng/g for cyclodienes (all as weight concentration in whole body tissues). The levels recommended by the Swedish Food Regulation are 5000 ng/g for DDTs, 2000 ng/g for PCBs, and 200 ng/g for HCHs [36]. The US Food and Drug Administration (FDA) tolerance limit is 2000 ng/g; wet weight for total PCBs in fish and shellfish [28] [37]. All these levels were higher than those recorded in the present study either in *Sparus auratus* recorded in both of the Eastern and the Western sectors.



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