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Organochlorine Pesticides and Polychlorinated Biphenyls Carcinogens Residual in some Fish and Shell Fish of Yemen

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Abstract

The concentrations of Dichloro-Diphenyl Trichloroethane (DDTs) as organochlorine pesticides (OCPs) and polychlorinated biphenyls (PCBs) in seventeen important commercial species Fish and Shell Fish from the Gulf of Adan and Red Sea of Yemen Coast were investigated in this study. Relation between weight and length of the organisms and the values of DDTs and PCBs were also considered. The concentrations of DDTs and PCBs were measured, using GC chromatography and using electron capture dtector. The values of DDTs in the Red Sea ranged from 0.9-7.8 ng g^{-1} and the OCPs ranged from 0.1-1.0 ng g^{-1}. As for the fishes and shell fishes of the Gulf of Aden, the concentration ranges were 0.3-6.4 ng g^{-1} and 0.1-0.9 ng g^{-1} for DDTs and PCBs respectively. Although PCBs are not manufactured in Yemen, their presence can be attributed to the industrial usage and the possible dumping of some products which contain PCBs. The study of relationships between DDTs and PCBs and weights or lengths indicated that these compounds are not concentrated in the tissues of fish and shell fish species.

الملخص

في هذا العمل تم دراسة تراكيز المبيدات العضوية المكلورة والبوليمرات ثنائية الفنيل في سبعة عشر نوع من الأسماك الاقتصادية المهمة من مياه خليج عدن وساحل البحر الأحمر اليمني. كما درست العلاقة بين طول وزن كل سمكة الى هذه التراكيز.

تم قياس هذه التراكيز بواسطة جهاز الجاز كروماتوجرافي، فيم المبيدات العضوية المكلورة في اسماك البحر الأحمر كانت من 0.9 ألى 7.8 نانو جرام / جرام ، قيم ثنائية الفنيل عديدة الكلور من 0.1 الى 1.0 نانو جرام / جرام.

فيم المبيدات العضوية المكلورة في اسماك خليج عدن كانت من 0.3 الى 6.4 نانو جرام / جرام ، قيم ثنائية الفنيل عديدة الكلور من 0.1 الى 0.9 نانو جرام / جرام. وبالرغم من أن ثنايئة الفنيل عديدة الكلور لا ينتج في اليمن لكنه يتواجد في الاستعمالات الصناعية. هذه الدراسة أوضحت أن تراكيز هذه المركبات ليست بالكبيرة في أنسجة هذه الاسماك

Keywords: Organochlorine Pesticide; DDTs; PCBs; Fish; Shell fish; Gas Chromatography; Red Sea; Gulf of Aden

1. Introduction

Organochlorine pesticides are a class of toxic compounds characterized by their relative chemical and biological stability, and hence persistence in the environments. Consequently, organochlorine pesticides have been placed on the top of the list of potential environmental hazards. The persistent and widespread occurrences of organochlorine pesticides have stimulated research into the nature, behavior, fate of pesticides in addition to their metabolites in the environment. The widespread occurrences of these compounds as environmental pollutants have been reported for all major terrestrial, fresh water, and marine environments. Damage to marine environment, by organochlorine pesticides, is well documented. However, very few data are available for the concentration of chlorinated pesticides in the seas around India (Jackson *et al.,* 1994; McCain and Varanasi, 1991; Nicholson, 1980; Tanbe and Tatsukawa, 1991; Mansour, 2004; Pfeuffer and Rand, 2004; Chou and Lee, 2005). As India is predominantly an agricultural country, large quantities of pesticides (55,000 t) have been used in agriculture for many years (McCain *et al.*, 1992).

At present, almost nothing is known about the existence of these pesticides in the Red Sea and Gulf of Aden. Organochlorine pesticides (OCPs) including Dichloro-Diphenyl-Trichloroethane (DDTs) have been used in Yemen and neighboring countries for more than four decades. According to a survey conducted in March 1990, there were a total of 80 pesticides brands of which twenty four are not recommended or even outlawed (Al-Ghashm, 1991; Bidleman and Leonard, 1982). Shipboard measurements in the Indian Ocean were made in the northern Arabian Sea, Arabian Gulf and the Red Sea. Average DDTs levels in the Arabian Sea -Arabian Gulf - Red Sea were 25-40 times as much of what is found in North Atlantic background value. They have attributed these higher levels to the continued use of DDTs in countries bordering these areas (Farrington *et al.,* 1983).

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Red Sea is an extremely isolated body of water. Water losses through evaporation far exceed rainfall, run-off from rivers is negligible; and apart from the Suez Canal, the only other mean of water exchange with other oceans is through the straits of Bab-el- Mandab. The Yemen coast is characterized by a narrow coastal plain between the Gulf of Aden and the mountain range that parallels the shoreline. This range averages 1.070 m in height and influences the local weather, especially the wind. The Gulf of Aden, which connects the Red Sea with the Arabian Sea (Indian Ocean), is approximately 900 km long, and large rivers flowed from the interior to the sea. Today, small wadi channels have been eroded into older and, larger river channel (DouAbul and Al-Shwafi, 2000; Al-Shwafi, 2008; Abdallah, 1996; Edwards and Head, 1987).

The need to identify organic and inorganic pollutants in the Red Sea has become a major concern for all countries in the region within the past few years because of the need for baseline data (or background levels), the chronic pollution from industrial and anthropogenic sources and the continual pollution of the areas fishing ground. In addition, and because of the potential impact on marine life and fisheries, it is also important to know the extent of the pollution and its impact on marine life, and the period of the pollution impact. Hence, the first necessary step is the determination of the seriousness of pesticides in the marine environment including the magnitude of their residues. However, such data are not available for the Red Sea and Gulf of Aden. Therefore, the present study is aimed mainly at determining DDTs and PCBs residues in fish and shell fish, and to attempt to identify their major sources and to perform a baseline study on the pollution in the muscle tissues of the commercial fish species, which were surveying in the Red Sea of Yemen and Gulf of Aden. The main objectives of the present study are to determine the residue levels of potential pollutants mainly the DDTs and PCBs in the flesh of the seventeen fish and shell fish species collected from the Red Sea of Yemen and The Gulf of Aden, and to identify the most important sources of contamination to the region. Also this study will focus on the sub-lethal effects of the most prominent contaminants, as well as their possible implications with humans who consume them. In addition, to generate baseline data for further follow- up study in the region.

2. Material and Method

2.1. Sampling

Fish and shell fish samples were collected from several locations along the Red Sea and Gulf of Aden Coast of Yemen during summer 1998. The collection of samples depending on the feasibility and importance of the site itself. Seventeen species of fish and shell fish were collected from the local commercial fishermen of Aden City and Hodiedah City. Samples were collected by fishermen, and then they were frozen upon return to the city center. Samples of fish and shell fish, having similar size (length and weight) were chosen for each species. Sub-samples (5gm each) of 17 species were dissected according to the procedure described by (ROPME Manual, 1983). The edible portions (muscles) were only taken for the determinations of DDTs and PCBs analysis.

2.2. Chemical Analysis

All solvents were redistilled, using distillation apparatus with a 150 cm vacuum jacketed fractionation column filled with 3 mm diameter glass helices. DDTs and PCBs were determined using a Hewlett Pachard HP5890-GC with split/splitless injector and a 25 m x 0.3 mm fused silica capillary with a chemically bonded gum phase SE54 sodium chloride and sodium sulfate that were kiln fired at 450^{o} C overnight and cooled in a greaseless desiccator. Silica gel, used for column chromatography, was solvent extracted with n-hexane in a flass cartridge inserted into an extraction apparatus, as described by Ehrhardt (1987). After extraction, the silica gel was first dried in the same cartridge by passing ultra-pure nitrogen through; and was then activated by heating the cartridge in an electric tube oven to 200^{o} C for 6 h with nitrogen stream reduced to a few ml per minute.

The extraction method was based upon that of Wade *et al.* (1988). A total 5g of dried tissues was Soxhlet-extracted with methyl chloride and concentrated in Kuderna-Danish tubes. The extracts were fractionated by alumina: silic gel (80-100 mesh) chromatography. The extracts were sequentially eluted from the column with 50 ml of pentane (aliphatic fraction) and 200 ml of 1:1 pentane-dichloromethane (PCBs/DDTs fraction) and concentrated for GC analysis. The recovery rate exceeded 80% for all the measured samples.

DDTs and PCBs were separated by gas chromatography in the split/splitless mode, using an electron capture detector (ECD). A 30 m x 0.32 mm i.d. fused-silica capillary column was used for this purpose with a chemically bonded gum phase SE54 (J&W Scientific, Inc.) provided component separations. Four-calibration solutions were used to generate a nonlinear calibration curve. A sample, containing only PCBs, was used to confirm the identification of each PCBs congener. The surrogates DBOFB (dibromooctafluorobiphenyl), PCB-103 and PCB-198 were added during the extraction during the extraction of DDTs and PCBs. The internal standard, TCMX (tetrachloro-*m*-xylene), was added prior to GC/ECD analysis. The chromatographic programm for DDTs and PCBs analysis was 100^{o}C for 1 min, then 5^{o}C min $^{-1}$ until 140^{o} C, hold for 1 min, then 1.5^{o}C min^{-1} to 250^{o}C, hold for 1 min, and then 10^{o}C min^{-1} to a final temperature of 300^{o} C, which was held for 5 min. The detection limit for the method is 0.001 ng g^{-1}.

3. Result

Table 1 shows the average concentrations of DDTs and PCBs in different fish and shell fish species collected from the Red Sea of Yemen. The concentrations were averaged across at least three measurements. The results of DDTs revealed that the concentrations ranged from 0.9-7.8 ng g^{-1} dry weight with an average of about 3.5 ng g^{-1}. DDTs values showed no relation either with weight or with length. The PCBs ranged from 0.1 to 1.0 ng g^{-1} dry weight with an average of about 0.5 ng g^{-1}. Also here, no

Table 1. Number, weight, and length of the collected fish and shell fish species in addition to concentrations of DDTs (ng g^{-1}) and PCBs (ng g^{-1}) in the muscles of the fishes and shell fishes collected from the Red Sea of Yemen. The errors are collected from standard deviations.

Fish and shell fish species	No	Total weight (g)			
Scomberomorus commerson	5	250-500	80-100	5.3±0.03	0.3± 0.04
Crenidens crenidens	15	500-600	30-50	4.2±0.04	0.5±0.02
Rastrelliger kanagurta	20	50-65	22-30	7.8±0.05	0.7±0.03
Thunnus albacares	7	500-700	70-90	3.9±0.07	0.4±0.02
Carchaias palasrras	10	200-500	55-62	2.4±0.03	0.1±0.01
Himantura uarnak	10	255-450	62-70	6.5±0.06	0.9±0.05
Caranx sem	10	105-320	50-55	6.7±0.09	0.4±0.02
Scomberoides commersonianus	10	562-765	72-75	5.1±0.03	0.3±0.03
Chanos chanos	10	50-108	35-40	4.8±0.07	0.5±0.02
Lutjanus sanguineus	10	120-142	34-47	1.2±0.07	0.8±0.06
Rachycentron canadus	8	152-256	60-63	2.0±0.03	0.9±0.05
Euthynnus affinis	10	320-450	44-50	2.8±0.02	0.7±0.07
Epinephelus areolatus	10	280-320	35-37	1.9±0.01	1.0±0.01
Panutirus homarun	10	320-460	40-42	2.3±0.03	0.2±0.05
Sepia pharnais	15	268-357	33-40	1.8±0.02	0.4±0.03
Sphyraena jello	10	365-452	43-52	1.1±0.01	0.3±0.04
Penaeus semisulcatus	25	95-120	25-30	0.9±0.01	0.1±0.02

Table 2. Number, weight, and length of the collected fish and shell fish species in addition to concentrations of DDTs (ng g^{-1}) and PCBs (ng g^{-1}) in the muscles of the fishes and shell fishes collected from the Adan Gulf. The errors are collected from standard deviations.

Fish and shell fish species	No	Total weight (g)	Total length (cm)	DDTs (ng g-1)	PCBs (ng g-1)
Scomberomorus commerson	5	270-450	80-90	4.3±0.02	0.2±0.02
Crenidens crenidens	15	620-650	33-54	2.8±0.03	0.3±0.01
Rastrelliger kanagurta	20	70-80	32-73	5.3±0.04	0.4±0.02
Thunnus albacares	7	453-720	60-89	2.8±0.05	0.2±0.01
Carchaias palasrras	10	500-700	57-65	2.3±0.03	0.1±0.01
Himantura uarnak	10	259-470	65-73	5.3±0.03	0.7±0.03
Caranx sem	10	115-325	53-59	6.4±0.07	0.5±0.02
Scomberoides commersonianus	10	653-777	75-77	5.0±0.02	0.4±0.02
Chanos chanos	10	55-112	38-44	4.3±0.06	0.3±0.03
Lutjanus sanguineus	10	120-142	36-50	1.0±0.03	0.6±0.04
Rachycentron canadus	8	170-268	66-69	1.7±0.02	0.8±0.03
Euthynnus affinis	10	333-470	49-53	2.5±0.02	0.6±0.06
Epinephelus areolatus	10	295-354	38-40	1.0±0.01	0.9±0.01
Panutirus homarun	10	332-472	43-45	2.3±0.03	0.3±0.03
Sepia pharnais	15	297-380	38-45	1.5±0.02	0.4±0.02
Sphyraena jello	10	395-460	46-55	1.0±0.01	0.2±0.03
Penaeus semisulcatus	25	100-129	35-43	0.3±0.01	0.1±0.01

relationship between PCBs concentrations and weight or length was observed. The results of DDTs and PCBs in the fishes of the Gulf of Adan are shown in table 2. The ranges of DDTs extended from 0.3 to 6.3 ng g^{-1} with an average of 2.9 ng g^{-1} and the ranges of PCBs extended from 0.1 to 0.9 ng g^{-1} with an average of 0.4 ng g^{-1}. No relationship was found between weight or length and DDTs and PCBs (Fig. 1 & 2). Statistical analysis of the relations shows r^2 values less than 0.1. Generally, the concentrations of both DDTs and PCBs in the fishes and shell fishes collected from the Red Sea of Yemen were higher than those collected from the Gulf of Adan. However, statistical analysis of the data using t-test shows that the differences were not significant either in DDTs (P= 0.1331) or in the PCBs (P=0.2313).

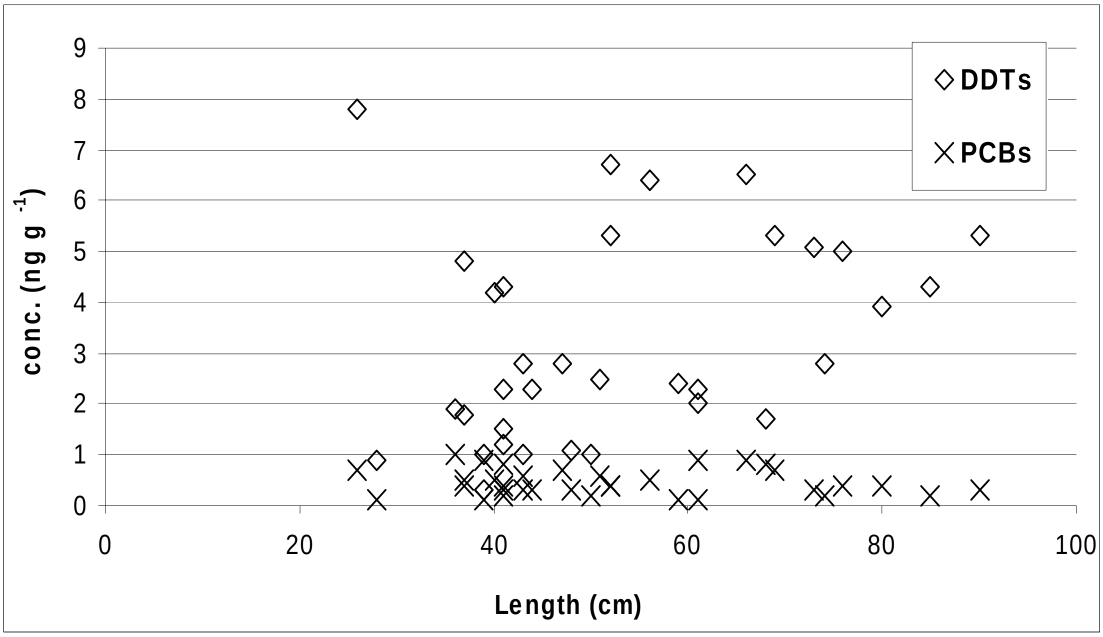

Figure 1. Relations between length (cm) and DDTs and PCBs concentrations (ng g^{-1}).

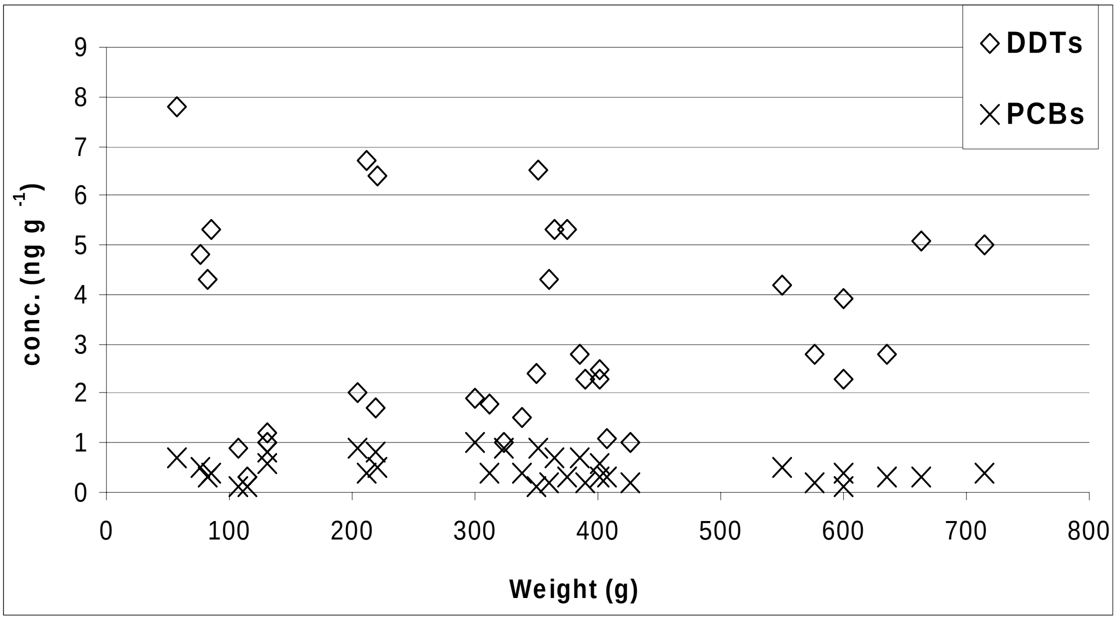

Figure 2. Relations between weight (g) and DDTs and PCBs concentrations (ng g^{-1}).

4. Discussion

It is difficult to compare the present results with other results from other studies since little information is available, especially in the Arabian context. However, DDTs in fishes and shell fishes from the Red Sea and Gulf of Aden were an order of magnitude lower than the range of values reported for fishes from the eastern Arabian Sea (Shailaja and Sen, 1989; Kishimba *et al.*, 2004). Similarly DDTs, determined in the present work, was an order of magnitude lower than the range of values reported for *Epinephelus tauvina* and *Lethrinus nebuluosus* captured from the coastal waters of Oman (Burns *et al.*, 1982). One of the primary concerns regarding DDTs is its potential for affecting fish reproduction. Early studies with trout demonstrated that DDTs concentrations in eggs or larvae in the ppm range impaired viability (Allison *et al.*, 1964; Hogan and Brauhn, 1975; Arambarri *et al.*, 2003). A number of dialed studies have also suggested that egg and larval viability or larval fitness were affected when relatively high levels of organochlorines were present in ovary tissues (Nelson *et al.*, 1991; Arambarri *et al.*, 2003; Mansourm and Sidky, 2003). Similar studies investigating DDTs and PCBs were done also in Kuwait waters and Abu Dhabi waters by DouAbul *et al.* (1987a). The authors found concentrations of 8.8-88 and 30 ng g^{-1} for DDTs and values of 5 and 7 ng g^{-1} for PCBs in Kuwait and Abu Dhabi water respectively. These values are much higher than the values reported in this study.

The higher concentrations of both DDTs and PCBs in the fishes and shell fishes collected from the Red Sea of Yemen, compared to those collected from the Gulf of Adan, may be attributed to the use of these compounds for agriculture in high mountains, where summer monsoon fall in Yemen and the water flowing mainly into the Red Sea and the Gulf of Adan. However, the amount of water flowing into the red sea is higher than the water flowing into the Gulf of Adan, which causes these higher concentrations. In addition, Red Sea is a small and semi-enclosed body when compared to the Gulf of Adan.

Polychlorinated biphenyls (PCBs) are a class of synthetic, inert, and complex mixture of many compounds originally manufactured in the USA in 1929, and until recently retained in large quantities mainly as dielectric fluids in electrical equipment (Hutzinger *et al.,* 1974; Bleatchy, 1984). Additionally, PCBs are extremely persistent in the environment, and like many other organochlorines of similar nature are widely spread among living organisms all over the world (Tanabe and Tatsudawa, 1991; Koeman and Visser, 1992).

Organochlorine compounds are generally soluble in fatty tissues, but exhibit low solubility in water. This lipophilic hydrophobic characteristic is largely responsible for their bioaccumulation and extreme persistence in marine biota (DouAbul *et al.*, 1987 a). The multiresidual extraction procedure employed in the present work followed by GC-electron capture determination should screen most of the common OCPs and PCBs in all spices. In most cases, conversion of DDT into DDE is initiated by soil micro-organisms immediately after it enters the environment. Other factors such as alkaline pH, light, or heat may also produce chemical changes in the original DDT molecule. Thus, the above observation may be due to metabolic conversion (Bridges *et al.*, 1963) and/or dehydrochlorination (Hannon *et al.*, 1970; DouAbul *et al.,* 1988; DouAbul and Heba, 1995) in the warm, rather alkaline waters of the Red Sea and Gulf of Aden. Moreover, because the volatility of DDE is several times greater than that of DDT (DouAbul *et al.*, 1987 b). It is logical to presume that DDE is more readily transferred via atmosphere to the coastal Red Sea and Gulf of Aden. Since the presence of the original compound suggests recent inputs of DDT to aquatic ecosystem (Aguillar, 1984). It may thus be concluded that there was continuing contribution of DDT to the Red Sea and Gulf of Aden. However, DDT has been officially banned in Yemen. Hence, its residues must be originated from a more remote source or more likely, from continuing illegal use. Technical DDT generally contains <25% *o.p*-DDT, an impurity, however, *o.p*-isomers are less persistent than their *p.p*-analogs (Fry and Toone, 1981; Mansour and Sidky, 2003). This phenomenon may be accounted for the relatively low percentage occurrence of both *o.p*-DDT and *o.p*-DDD, which were 10% and 0% respectively.

Chlorinated pesticide and PCBs residues in commercial fishes caused leukemia cancer, gastric carcinoma, and esophageal, (Haffer, 1983; 1985). PCBs are not manufactured in Yemen, and their presence can thus only be from industrial usage and the possible dumping of products containing PCBs. To the best of the author's knowledge, previous data for PCBs in the Red Sea and Gulf of Aden are not available. The present results are the first of their kind for the region and should serve as background information.

The major conclusions that can be drawn from the present study are; i) the concentrations of the most organochlorine residues in the commercial fish of the Red Sea and Gulf of Aden environment were either below the detection limits or in very low values ii) DDTs occurred in almost all fish samples examined. However, DDTs concentrations were relatively lower than those reported previously in the Arabian Sea. iii) traces of PCBs were also found in some samples of fishes and shell fishes from the Red Sea and Gulf of Aden.

Generally, it is recommended that a continuos monitoring programme for the Red Sea and Gulf of Aden region should be formulated and conducted to ensure that the concentration of DDTs and PCBs are within the base line levels established in the present study.

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