Bull. Environ. Contam. Toxicol. (2004) 73:306–311

DOI: 10.1007/s00128-004-0428-7

Bulletin of Environmental Contamination and Toxicology

Accumulation of Organochlorine Pesticides in Marine Fishes from Coast of Taoyuan in Taiwan

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Received: 3 October 2003/Accepted: 13 May 2004

Organochlorine compounds have long been recognized as the most deleterious contaminants to biota in the world's marine and estuarine waters. The concentration of DDT and its metabolites increase through successive trophic levels by more than 1000-fold, for example, from 0.04 mg/kg in plankton to 75 mg/kg in a Ring-billed Gull (Murty, 1988). These compounds are deliberately added to the environment for the purpose of killing or injuring certain life forms, and because of that, they have a rather unique position among chemicals that humans encounter daily. Most of the chemicals used as pesticides are not highly selective but are generally toxic to other species, including humans and other desirable forms of life that share the environment.

In Taiwan, the climate is characterized by high temperatures and humidity, which is suitable for breeding fit pests; thus a large number of organochlorine pesticides were used for pest control in agriculture and for prevention and cures for public hygiene. From 1950 to 1974, organochlorine pesticides were generally used on farmland and in the environment; after that time, multiple residual and toxicity problems with organochlorine pesticides were found. Bans on DDT, BHC, lindane, aldrin, endrin, dieldrin, and heptachlor were later enacted (Liao, 1984; TACTSRI, 1993). During the past 20 years, various biomonitoring strategies have been developed to monitor and evaluate the adverse impacts of these compounds on marine ecosystems in Taiwan. Han et al. (2000) reported that mussels/bivalves were good indicators for *p,p* -DDT, *p,p* -DDD, and *p,p* -DDE, but a drawback is that mussels are not found everywhere. Ling et al. (1994) reported the average daily fish consumption of PCDD/PCDFs will exceed the accepted level of risk. Therefore, marine fishes are the one of the bioindicators which cannot be ignored.

The objective of this work was to study the presence and concentrations of organochlorine pesticide residues in marine fishes from the coast of Taoyuan, northwestern Taiwan, an area with freshwater, waste agricultural, and industrial water inputs.

MATERIALS AND METHODS

Fish were collected from the Taoyuan coast (Fig. 1) using cast nets during July and October 1999. Immediately after collection, the fish were stored on ice in an ice bucked and transported to the laboratory and stored -20℃ until dissection. We selected species of fish which had been collected at most stations as samples for analysis.

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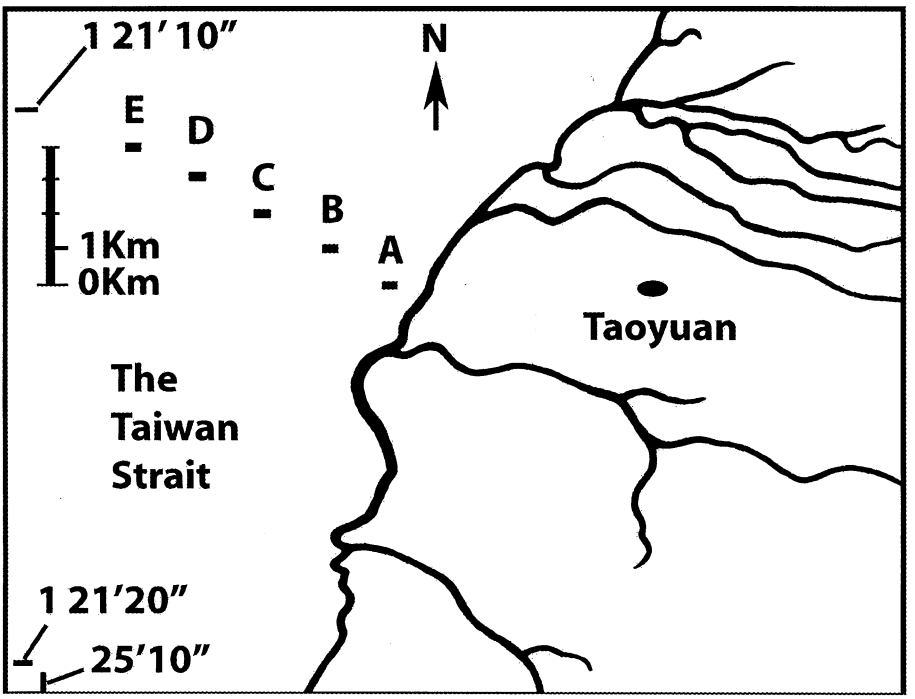

Figure 1. Sampling locations (A, B, C, D, E) of fish collected from Taoyuan sea area in Taiwan.

Prior to analysis, the liver and muscle tissues of one to ten fish from each station were dissected out, according to the number of fish caught. Liver and muscle tissue samples were analyzed by mixing tissue samples of the same station. Sample preparation and analysis were carried out according to the procedures described by the Official Methods of Analysis (1970) of the AOAC (Association of Official Analytical Chemists, US). Samples were freeze-dried before analysis; approximately 2 g (dry weight) was extracted with 10 g of anhydrous sodium sulfate and 50 ml benzene during 20 min in a mechanical shaker (200 rpm). After concentration to 2~3 ml, the extracted solution was passed through a purifier which contained 10 g Florisil powder and 2 g anhydrous sodium sulfate. Then a rotary vacuum evaporator was used to dry the material, and finally 1 ml isooctane was added. The organochlorine pesticides, including lindane, aldrin, dieldren, endrin, heptachlor, heptachlor epoxide, *p,p* -DDT, *p,p* -DDD, and *p,p* -DDE, were separated and quantified by an HP 5890 series II plus gas chromatograph equipped with an HP 5989B MS ENGINE mass spectrometer, electronic pressure control, and a capillary column (DB-5 MS, 30 m x 0.25 mm, 0.25 μm, J&W Scientific Products). The identification of compounds was deduced from their retention times, and quantification was based on peak height/area measurements as well as comparisons with responses of reference standards. Blanks were run for the entire procedure, and blank corrections were applied for each set of analyses.

RESULTS AND DISCUSSION

The species of fish found at the five stations are given in Table 1. Only one

Table 1. Species of fish and their number found at the five station.

Fish species/	July			October				
Station no.	A	B	C	A	B	C	D	E
Megalops cyprinoids	0	4	2	0	0	0	1	0
Sardinella sindensis	0	1	0	0	0	0	0	0
Pennahia pawak	2	2	0	0	0	1	0	0
Terapon jarbua	0	1	0	0	0	0	0	0
Pomadasys hasta	2	2	3	3	4	4	5	5
Arius maculates	0	0	2	1	2	1	0	0
Trichiurus lepturus	0	0	0	0	0	0	3	0
Epinephelus amblycephalus	0	0	0	0	0	1	0	0
Epinephelus fasciatus	0	0	0	0	2	0	0	0

species appeared at all five stations, *Pomadasys hasta*. Because adverse weather conditions suddenly occurred while we were on the ocean, we were only able to complete sampling at three stations in the first of July.

Concentrations of organochlorine compounds by month and station are given in Tables 2 and 3. Only *p,p* -DDD and *p,p* -DDE were found in the muscle and liver of sampled fish. The highest median concentration in muscle for *p,p* -DDD was 488.33 μg/L in July and 104.35μg/L for *p,p* -DDE in October. The highest median concentration in the liver for *p,p* -DDD was 796.32 μg/L in July and was 465.62 μg/L for *p,p* -DDE in October. The lowest concentrations of *p,p* -DDD and *p,p* -DDE appeared at station B, and no *p,p* -DDE was found in July and October.

The aquatic organisms studied showed an accumulation of pesticides, with the highest occurrence occurring for *p,p* -DDD followed by *p,p* -DDE. Concentrations of *p,p* -DDD at stations A and B increased, and at stations B and C decreased in July. The same situation occurred in October, with an increase at stations A to D, then a decrease from stations D to E.

Over 20 years ago, the government had forbidden the use of organochlorine compounds. Jen and Sun (1974) reported that aldrin, eldrin, heptachlor, and heptachlor epoxide were not detected, including in cultured fishes, mussels, and bivalves in Tainan, Yunlin, and other areas. Among the isomers of DDT, *p,p* -DDD was the highest (with a mean of 14 μg/L) followed by *p,p* -DDE (with a mean of 8 μg/L), and the lowest was *p,p* -DDT (with a mean of 7 μg/L). These results were similar to findings of our research, revealing that most residues of *p,p* -DDD and *p,p* -DDE are the degradation products of *p,p* -DDT.

The main mechanism for DDT's metabolism is a dechlorination reaction, through which mechanism DDT is transferred to DDD (Liao, 1984); this mostly occurs under anaerobic conditions by microorganisms. In 1972, Patil et al. separated 100 lines of microorganisms from surface water membranes and sediments of seawater, and found 35 lines which could degrade *p,p* -DDT after testing; the

Table 2. Organochlorine compounds found in fish samples from the coast of Taoyuan, including concentrations in muscle (μg/kg) and liver (μg/kg) at five stations in July 1999.

Organochlorine compounds	Muscle			Liver		
	A	B	C	A	B	C
Lindane	ND	ND	ND	ND	ND	ND
p,p'-DDD	80.01	488.33	289.53	251.32	796.32	355.01
p,p'-DDE	99.11	ND	95.38	310.03	ND	266.05
p,p'-DDT	ND	ND	ND	ND	ND	ND
Aldrin	ND	ND	ND	ND	ND	ND
Dieldren	ND	ND	ND	ND	ND	ND
Eldrin	ND	ND	ND	ND	ND	ND
Heptachlor	ND	ND	ND	ND	ND	ND
Heptachlor epoxide	ND	ND	ND	ND	ND	ND

ND=Not detected

Table 3. Organochlorine compounds found in fish samples from the coast of Taoyuan, including concentrations in muscle (μg/kg) and liver (μg/kg) at five stations in October 1999.

Organochlorine compounds	Muscle					Liver				
	A	B	C	D	E	A	B	C	D	E
Lindane	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND
p,p'-DDD	73.33	79.25	81.62	190.12	91.81	431.62	434.52	459.53	653.31	175.35
p,p'-DDE	91.85	ND	93.67	104.35	98.72	453	ND	465.62	214.35	201.15
p,p'-DDT	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND
Aldrin	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND
Dieldren	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND
Eldrin	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND
Heptachlor	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND
Heptachlor epoxide	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND

ND=Not detected

major relative was *p,p'*-DDD. After treatment by a solvent, it was found that a large portion of the labeled substances of the water phase remained, and this would explain why some DDT relatives are soluble in water (Patil, 1972). At the same time, they found that some algae and plankton could also transfer *p,p'*-DDT to *p,p'*-DDD, and some papers reported that phytoplankton could transfer *p,p'*-DDT to *p,p'*-DDE in a 9-day experimental period; the ratio of DDT transferred increased from a trace to about 7.5%. According to another part of this research project, we knew that there were at least two diatoms, *Skeletonema costatum* and *Nitzchia* sp., which could absorb and metabolize DDT, with the resultant product being entirely DDE. That was found at all stations except B. A previous investigation revealed that the mean total DDT in the water of the Tanshui River was 0.014 μg/L between 1973 and 1976 (Ku, 1979), which was higher than the value 0.0068 μg/L of water in the Tatu River. An investigation by the Taiwanese EPA found from 1998 to 1999 (Chen, 1999), after analyzing 36 river water samples, that the rate of detection of many of the banned organochlorine pesticides in the land-river system had decreased.

Since organochlorine pesticides, such as DDT, are known to spread far and wide because of their resistance to decomposition and their lipophilic properties; the global monitoring of these pesticides has become one of the world's most important priorities (Kuehl, 1994; Marquis et al., 1994; Marquis et al., 1994; Muty, 1986). In Taiwan, organochlorine pesticides have been decreasing, and are gradually being replaced by organophosphates, carbamates, and synthesized pyrethroids. Therefore, to integrate the analysis and investigation results in this study, which reveal that land regions, including the soil, freshwater, and general organic tissues, have decreased contents of organochlorine pesticides, then we can believe that the government policy banning these substances worked. In the ocean systems of Taiwan, previous data were not plentiful, with only Han et al. (2000) reporting that the total content of *p,p'*-DDT was higher at 340 ng/g (dry weight) in mussels/bivalves in the Matsu marine area, but it was only 34.9 and 6.2 ng/g (dry weight) in Anping and Penghu. So, our study shows that on the Taoyuan coast, an important level of pollution by organochlorine pesticides still exists, and some of these chemicals are present in concentrations higher than that seen in other places, even though their use is forbidden by law. In conclusion, we suggest that it is imperative to implement stricter environmental controls in coastal areas in order to minimize the potential risks of these substances to other life forms including humans.

Acknowledgments. We thank the Hui-Qun Technology Co. for their financial support of this study. We also thank Dr. Chen (Professor of the Department of Zoology of National Taiwan University) for help with revision of the paper.

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