

LEVELS OF 18 POLYCHLORINATED BIPHENYL CONGENERS IN FIVE FISH SPECIES IN TAIWAN

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Introduction

Traces of Polychlorinated biphenyls (PCBs) have been reported in seafood, vegetables, and other food stuff throughout the world¹⁻³. Seafood is thought to be the major source of PCB exposure to general public. Pan et al. analyzed PCBs in fishes and dairy products in Taiwan and reported that there were traces of PCBs in fish, meat and dairy products⁴. However, the analysis was not conducted in a congener-specific basis. Since different congeners have different chemical and physical property, they also have different toxicity⁵. Congener-specific analysis is important to further evaluate the potential health effects resulted from this route of exposure.

Yu-Cheng incidence in 1979 in Taiwan had called attention to PCB poisoning. In the follow-up epidemiological study, not only Yu-Cheng women but also controls have PCB concentrations in their bloods. The ratio of PCB concentrations in serum of Yu-Cheng women to those of controls was the lowest compared to PCDDs and PCDFs⁶. The extent of possible exposure of general public in Taiwan from daily intakes of fishes, dairy products and other food supply is worth investigation. This work analyzed eighteen PCB congeners for samples of five fish species obtained at major markets in Taiwan in order to assess potential PCB intakes of general public from fish consumption.

Methods and Materials

Five fish species were selected according to the consumption quantity in Taiwan⁷, including Tilapia (*Oreochromis hybrids*), Milkfish (*Chanos chanos*), White pomfret (*Pampus argenteus*), Hairtail (*Trichiurus lipturus*), and Cod (*Theragra chalcogramma*). They are all highly-consumed species; Tilapia is the most popular one through out Taiwan Island. Tilapia and Milkfish are both from cultivated fresh-water or salt-water ponds. Both White pomfret and Hairtail are caught along the west coast of Taiwan. Cod is imported from other countries.

Approximately ten fish samples of each species were randomly purchased at the major markets in each of three major cities in Taiwan: Taipei, Taichung, and Kaohsiung in the northern, central, and southern Taiwan, respectively. They are the main consumer markets in Taiwan. Purchased fishes were refrigerated at -20°C; they were thawed before sample preparation. Approximately 10 grams of minced fillets was put in pear-shaped glassware and extracted with 80mL hexane/acetone (volume ratio 8:2) mixtures by Tissuemizer three times.

Kuderna-Danish (K-D) apparatus was used to concentrate the extracts to about 1 mL, which was then cleaned up by 6g of 2% deactivated silicon gel and 3g of 3% deactivated alumina oxides with hexane. Elutes were concentrated down to about 1 mL under a gentle stream of nitrogen and

transferred to GC vials. PCB analysis was performed on a Perkin Elmer M611-9000 Gas Chromatography-Electron Capture Detector (Perkin Elmer Corporation, Norwalk, CT), equipped with a 30m× 0.25mm ID× 0.25µm Perkin Elmer PE-5 capillary column. The temperatures of the injection port and the electron capture detector were 280°C and 300°C, respectively. Quantification of GC results was conducted by internal standard method.

Lipid contents of fishes were assessed in a similar way. Approximately 10 grams of minced fillets were put in pear-shaped glassware and extracted with 60mL hexane by Tissuemizer three times. Extracts were concentrated to about 1 mL by K-D apparatus, poured into pre-weighed aluminum cups, and baked at 80°C. Aluminum cups were weighed after 24, 48 and 72 hours till reaching stable weights to obtain fat contents.

Eighteen congeners were selected, including IUPAC No. 28/31, 52, 66, 77, 105/153, 118, 126, 128, 138, 156, 157, 167, 169, 170, 180 and 189. Analytical standards of individual PCB congener in hexane were purchased from Ultra Scientific, North Kingston, Rhode Island. The purity of all standards was 99% or higher.

There was one procedural blank for each batch of ten samples to assess any possible contamination. The levels of contamination of all congeners were below 1ng. One matrix spike sample per batch was conducted to assess percent recovery rates of different PCB congener. Totally, 15 matrix spike samples were analyzed, the recovery rates were from 70% to 110%. Surrogates (PCB IUPAC No. 103 and 112) were added to each sample to obtain percent recovery rates of each sample. They ranged from 70% to 130%. Internal standard PCB IUPAC No. 166 was added to each sample and used to quantify PCB congeners.

Concentrations of PCB congeners were expressed as wet weight concentrations (ng/g wet) and lipid concentrations (ng/g lipid). The reported data were all blank-corrected, but not adjusted by percent recovery rates. Those concentrations below method detection limits (MDLs) were taken as 1/2MDL. Krustal-Wallis test and Scheffe test in Statistical Analysis Software (SAS version 6.12, SAS Institute Inc., Cary, NC) were used to compare PCB concentrations with different classification. Toxic equivalent quantities (TEQs) were calculated by multiplying concentrations of each congener by its toxic equivalent factor, which was from WHO (WHO-TEFs).

Results and Discussion

In total, 146 samples were analyzed. The lipid content of eatable parts were 1.2±1.1%, 5.1±2.8%, 0.89±0.61%, 1.1±1.3%, and 12.5±9.0% for Tilapia, Milkfish, White Pomfret, Hairtail, and cod samples, respectively. The measured lipid contents were comparable to those reported in the literature. Cod samples had the highest lipid content among those five fish species. There was no significant weight or lipid content difference among fishes from different markets.

Among 146 samples, 37%, 63%, 58%, 86%, and 100% of Tilapia, Milkfish, White pomfret, Hairtail, and Cod samples, respectively, were found to have one or more PCB congeners. PCB concentrations are summarized in Table 1. Cod samples had the highest wet weight concentrations of PCBs; they were statistically significantly higher than PCB concentrations of the other four species (p=0.0001). PCB concentrations in Hairtail samples were enhanced considerably after being adjusted by lipid content; they were statistically significantly higher than Tilapia, Milkfish, and White pomfret samples (p=0.0001).

Tilapia and Milkfish are cultivated only for less than one year before being sold on the market. The percentage of contaminated samples and PCB concentrations in those contaminated fishes were not high. White pomfret, Hairtail, and cod are demersal fishes in the ocean. Their life spans are longer; thus, they have more chance to accumulate contamination around them. The market-size white pomfret and Hairtail are about 2~4 years old. Cod samples were slices cut from originally much bigger bodies; the market-size cod could be 5-10 years old; and they had the highest wet weight concentrations of PCBs.

Chou et al. (1987) found that mean values of total PCBs in freshwater and seawater fish samples from Taiwan markets were 21.0 (1.1~269.3) and 41.1 (0~411.1) ng/g wet wt, respectively⁸. In 1991, Pan et al. reported that total PCB concentrations in fish samples had dropped to 0.2~16.1 ng/g wet wt⁴. This work showed that levels of 18 congeners in fish samples were about 0.92~45.2ng/g wet wt. Since the analytic methods were different among different studies, we cannot conclude whether the PCB concentrations in fish were decreasing over the years.

Congener profiles of studied five fish species are in Table 2. There was quite variation among samples of the same species. Nevertheless, PCB 138 was the major congener in Tilapia samples, while PCB 52, 66 and 77 accounted for the majority of PCBs in the other four species. PCB 126 had the highest TEF; samples with traces of PCB 126 such as Hairtail had highest TEQs (Table 1).

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Table 1 Concentrations and WHO-TEQs of 18 PCB congeners in five fish species (mean±SD)

Species	Tilapia	Milkfish	White Pomfret	Hairtail	Cod
(ng/g wet)	2.06±0.30	1.87±0.42	2.53±0.62	3.04±1.55	8.92±8.8
(ng/g lipid)	14.4±22.5	9.65 ±13.3	143.2±240.3	228.6±249.6	273.6±329.3
(pg-TEQ/g wet)	0	0.5±1	0±0.1	2.3±3.4	0.7±1.7

Table 2 Percentage of different PCB Congeners in Five Fish Species

PCB NO	Tilapia	Milkfish	White Pomfret	Hairtail	Cod
31/28	28.2%	23.6%	13.2%	6.2%	2.8%
52	0.0%	16.3%	42.9%	17.7%	17.7%
66	0.0%	35.6%	35.3%	14.2%	2.9%
77	0.0%	11.5%	8.5%	16.2%	3.4%
118	0.0%	2.4%	0.0%	4.3%	18.7%
105/153	4.3%	1.8%	0.2%	5.3%	17.8%
138	50.0%	1.3%	0.0%	8.5%	25.1%
126	0.0%	1.8%	0.0%	3.8%	0.0%
128	0.0%	0.0%	0.0%	0.0%	5.1%
167	0.0%	2.3%	0.0%	1.9%	0.1%
156	9.9%	0.1%	0.0%	0.7%	0.0%
157	5.0%	2.3%	0.0%	1.3%	0.0%
180	2.6%	0.8%	0.0%	6.3%	4.7%
169	0.0%	0.1%	0.0%	0.1%	0.0%
170	0.0%	0.1%	0.0%	4.5%	1.7%
189	0.0%	0.0%	0.0%	1.7%	0.2%