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Butyltin Compounds in Fishes Commonly Sold in Taiwan Markets

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ABSTRACT

Fishes commonly consumed in Taiwan were purchased from four fish markets at seasonal intervals in 2006 in order to monitor the butyltin residues, including monobutyltin (MBT), dibutyltin (DBT), tributyltin (TBT), and tetrabutyltin (TeBT), in fishes caught along the west coast of Taiwan. Concentration of MBT, DBT, TBT, and TeBT in the muscle or the internal organ of these fishes were in the range of n.d.–949.2 ± 49.1, n.d.–56.2 ± 7.9, n.d.–70.8 ± 10.0, and n.d.–103.8 ± 76.9 ng/g (wet weight), respectively. Owing to the toxicological similarity between DBT and TBT, total concentrations of DBT plus TBT in the muscle or the internal organ were taken as the standard for the residue level of butyltins and they were found lower than the tolerable average residue level (TARL) of 175.4 ng/g (wet weight). Therefore, butyltin levels in fish from Taiwanese markets are not a matter of concern for public health.

Key words: butyltin compounds, Taiwan, fish markets, tolerable average residue level (TARL)

INTRODUCTION

Over the last 40 years, organotin compounds have been extensively used as a pyrolysis stabilizer in the production of polyvinyl chloride (PVC), as a catalyst in the manufacture of polyurethane and silicone elastomer, as a stabilizer for improving resistance to ultraviolet radiation, and as a biocide to prevent fouling(1). Therefore, they are present in PVC food packaging materials, polyurethane foams, antifouling paints, agrochemicals, and many other consumer products.

Through leaching from the antifouling paints used in boat, ship and other marine equipments, from the PVC products disposed into sanitary landfills, as well as from the runoff of agricultural fields, these compounds have been introduced into our estuarine and marine ecosystems. As organotin compounds are bioaccumulative(1), tributyltin (TBT) and its breakdown products, monobutyltin (MBT) and dibutyltin (DBT) have been detected in a range of marine species including edible fish as well as invertebrates such as mollusks, crustaceans and cephalopods(1).

Organotin compounds have harmful effects on a variety of non-targeted organisms, even at low nanomolar aqueous concentrations(1,2). They are also suspected to have an endocrine disrupting ability in human. They may cause abnormalities in male reproductive systems(3,4,5) and disrupt the critical function of human immune cells, particularly the killer cells which fight infection. Recent findings have revealed biologically significant levels of organotins in random human blood samples from the USA, and in human liver from Poland(6).

Several investigations have shown that seafood is the primary source of human exposure to organotin compounds either in Asia or Europe(7,8,9). Through the consumption of contaminated seafood, organotin compounds may enter the human body.

In Taiwan the use of organotin compounds in antifouling agents was restricted to boats under 25 m in length in 2003. However, butyltin compounds were still encountered in water and sediments(10,11). This was attributed to TBT leached from antifouling paints from big commercial vessels (larger then 25 m in length) and/or the persistence of TBT in sediments(12).

Since many fishes consumed in Taiwan are harvested from the coast of Taiwan, it becomes necessary to monitor the levels of organotin compounds in fish commonly sold in markets in order to evaluate the extent of contamination after the controls of TBT, and to calculate the potential health risks to the public from consuming market-bought seafood. In this study, headspace solid-phase microextraction (SPME) and gas chromatography-flame photometric detection (GC-FPD) analytical procedures were used to quantitatively determine the butyltin compounds in fishes from four fish markets in Taiwan.

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MATERIALS AND METHODS

I. *Sample Collection*

Between March 2006 and November 2006, fish samples (about four to six fish species) were purchased from the fish markets in Keelung Bi-Sha (northern part of Taiwan), Hsinchu Nan-Liao (north central part of Taiwan), Taichung Wu-Gi (central part of Taiwan), and Pintung Dong-Gang (southern part of Taiwan), as shown in Figure 1, at seasonal intervals.

These fish samples included nine species of finfish (*Trichiurus lepturus*, *Psenopsis anomala*, *Dentex tumifrons*, *Polydactylus microstomus*, *Acanthopagrus berda*, *Nematalosa nasus*, *Psettodes erumei*, *Pampus argenteus*, and *Pennahia argentata*), and two species of shellfish (*Babylonia formosae* and *Lunella coronata*). They were all caught along the west coast of Taiwan. For each species three individual fish were purchased. The body length and body weight of the fishes were measured (Table 1). Subsamples were taken from the dorsal muscle of the finfish (about 20 g per subsample), the whole internal organ of the finfish (about 20 - 100 g per subsample), and the entire muscle of the shellfish (about 2 - 5 g per subsample) for analyses. Fish samples were frozen immediately after purchasing, and were transferred to the laboratory where they were stored at -20°C before analysis.

II. *Analytical Methods*

Butyltin compounds in various tissues of fish samples were analyzed as described previously[11,13]. The removed muscle and the internal organ were freeze-dried and homogenized before analysis. The analytical procedure consisted of five steps: (i) acid digestion of the dry tissue (1 g) with 10 mL of HCl: tetrahydrofuran (1:11) and homogenized in a Tekmar tissumizer; (ii) extraction with 30 mL of tropolone-benzene (0.1 g/L) twice in a shaker for 40 min; the two organic extract were then combined; (iii) SPME procedure[13,14]. In this procedure, solution containing 48.5 mL of artificial seawater, 1.5 mL of acetate buffer solution (pH 4.8), 50 μL aliquot of the fish extract, 250 μL of internal standard (tripropyltin chloride, 50 ng/L) and 100 μL of 1% sodium tetraethylborate ($NaBEt_4$, 98%; Strem Chemicals, France), were added into a bottle, and the fiber (100 μM polydimethylsiloxane; Supelco, USA) was then drawn into the bottle and situated approximately 1 cm above the aqueous surface. The bottle was incubated at 25°C for 50 min allowing for in situ $NaBEt_4$ derivatization and extraction to the fiber; (iv) the fiber retracted into the needle was immediately inserted into the GC injector for thermal desorption; (v) quantitative determination of butyltin compounds by Dani 1000 gas chromatographer equipped with a column (HP–5, 30 m × 0.25 mm i.d. × 0.25 μM film thickness, Hewlett Packard, USA) and a flame photometric detector fitted with a 610 nm optical filter. The fiber was injected in splitless mode. The column temperature was programmed from 70°C (1-min holding time) to 190°C at 30°C/min and to 270°C (3-min holding time) at 15°C/min, and then to a final 290°C (1-min holding time) at 15°C/min. The injector temperature was 250°C, and the detector temperature was 290°C. All experiments were conducted in triplicate. Recoveries of MBT, DBT, TBT, and tetrabutyltin (TeBT) from spiked-in fish tissue samples (0.3 μg/g, N = 6) were 94.0-95.4%, 98.2-111%, 101-110%, and 76.4-88.0% (w/w), respectively. Slight difference in recovery was observed among different fish species. The method detection limit of MBT, DBT, TBT and TeBT were 3.2, 3.8, 3.8, and 3.6 ng/g (dry weight), respectively.

III. *Crude Lipid Content Analysis*

Water content of the dorsal muscle or the internal organ of the finfish and the shellfish was measured based on the weight difference between the wet weight and the freeze-dried weight of these tissues, while crude lipid content in these tissues was determined by continuous petroleum ether extraction in a Soxtec system[15]. All experiments were conducted in triplicate.

Wet-weight-based butyltin concentration was calculated from the dry weight based butyltin concentration and the average water content in these tissues.

RESULTS AND DISCUSSION

I. *Biological Data of Samples*

As shown in Table 1 the average body length and body weight of *T. lepturus*, *P. anomala*, *D. tumifrons*, *P. microstomus*, *A. berda*, *N. nasus*, *P. erumi*, *P. argenteus*, *P. argentata, B. formosae* and *L. coronata* used in this study were in the range from 1.8 to 85.0 cm and 6 to 765 g, respectively (as show in Table 2-6).

Water content in the dorsal muscle ranged from 55.0 to 85.2% (w/w), as shown in Table 2-6, while the water content in the internal organs from 45.7 to 90.5% (w/w). Crude lipid content in the dorsal muscle of these fishes were in the range from 0.2 to 13.7% (w/w) and the crude lipid content in the internal organs from 0.8 to 38.9% (w/w).

II. *Concentration of Butyltin Compounds in Fish Samples*

Concentration of butyltin compounds in the fish samples collected from March 2006 to November 2006 are determined and shown in Table 2-6. Concentrations of TBT and the total concentration of butyltin compounds (BTs) in the muscle were in the range from n.d. to 70.8 ± 10.0 ng/g and n.d. to 953.7 ± 49.7 ng/g (wet weight), respectively (as show in Table 2-6). The highest concentrations of TBT (70.8 ± 10.0 ng/g) and BTs (953.7 ± 49.7 ng/g) were found in the muscle of *D. tumifrons* and *T.*

lepturus, respectively.

Concentrations of TBT and BTs in the internal organs were in the range from n.d. to 38.9 ± 9.8 ng/g and n.d. to 632.8 ± 76.0 ng/g (wet weight), respectively (as show in Table 2-6). The highest concentrations of TBT (38.9 ± 9.8 ng/g) and BTs (632.8 ± 76.0 ng/g) in internal organs were found in *T. lepturus* and *P. anomala*, respectively.

Although the concentration of each butyltin compound varied greatly for different individuals, different species, and different seasons, MBT was prevalent in most of the muscle and internal organ samples determined. Concentrations of MBT and BTs in the muscle and the internal organs of *T. lepturus*, *P. anomala, D. tumifrons* and *P. microstomus* were significant different ($p < 0.05$, by ANOVA test) from different months. We also found the mean of concentrations of MBT and BTs were higher in fishes caught in June (325.9 and 328.2 ng/g) than in the other months (64.8 and 72.0 ng/g, March; 91.8 and 106.4 ng/g, September; 88.5 and 108.0 ng/g, November), but there was no clear seasonal trend for other fish species. Different from our results, Dong *et al.*[10] found that ponyfish *Leiogenathus splendens* collected from the west coast of Taiwan contained higher BTs in winter than in other seasons.

In this study we also found that in some cases, concentrations of TBT or BTs were higher in the muscle, but in other cases, the concentrations of TBT or BTs were higher in the internal organs of the fish. Suzuki *et al.*[16] reported higher concentration of BTs in the liver than in the muscle of yellowtail. When determining butyltin compounds in the skin, dorsal muscle, ventral muscle, dark muscle, and liver in cobia *Rachycentron canadum* raised in offshore aquaculture sites, the researchers found that in most cases the highest concentration of total butyltin residues was in the liver or skin, but in some cases the highest concentration was in the muscle tissue[11]. Several factors such as temperature, diet, or enzyme activity that result in degradation of TBT in the tissues, might cause variation in the butyltin concentration in different tissues of the fish[17,18].

Concentrations of TBT and BTs in the muscle and the total length of *T. lepturus* (r = -0.249, r = 0.0141; N = 16), *P. anomala* (r = -0.443, r = 0.0714; N = 13), *D. tumifrons* (r = 0.112, r = 0.188; N = 11), and *P. microstomus* (r = -0.244, r = 0.105; N = 11) were not related. Concentrations of TBT and BTs in the internal organs and the total length of *T. lepturus* (r = 0.289, r = 0.199; N = 16), *P. anomala* (r = 0.265, r = -0.0693; N = 13), *D. tumifrons* (r = 0.127, r = 0.347; N = 11), and *P. microstomus* (r = -0.132, r = 0.290; N = 11) were also not related either.

No correlation was observed between the concentrations of TBT and BTs, and the lipid content in the muscle of *T. lepturus* (r = -0.155, r = 0.128; N = 14), *P. anomala* (r = 0.163, r = 0.191; N = 11), *D. tumifrons* (r = 0.544, r = 0.208; N = 10), and *P. microstomus* (r = -0.149, r = 0.101; N = 12). Neither was there a correlation observed between the concentrations of TBT and BTs, and the lipid content in the internal organs of *T. lepturus* (r = 0.202, r = 0.157; N = 12), *P. anomala* (r = -0.237, r = 0.720; N = 10), *D. tumifrons* (r = -0.371, r = -0.179; N = 8), and *P. microstomus* (r = -0.079, r = 0.267; N = 10). Harino *et al.*[19] also found that the concentration of TBT in muscle was not related to the total length of the fishes. Neither correlation was observed between the concentration of TBT and the lipid content in the muscle of fish.

Kannan *et al.*[6,20] surveyed the butyltin residues in the muscle tissue of fishes collected from several local markets and seafood shops in Asia and Oceanic countries between 1990 and 1994, and found that the total butyltin residues in tilapia, milkfish, and seabream bought in Taiwan were 0.49, 0.96, and 18 ng/g wet weight. On the other hand, a recent study has shown that TBT and total butyltin residue levels in the liver, dorsal muscle, ventral muscle and dark muscle of cobia (*R. canadum*) aquacultured in the offshore of Taiwan were n.d.–1140 and n.d.–52745, 75–338 and 79–688, n.d.–497 and 82–1715, and 66–528 and 93–803 ng/g (wet weight), respectively[11]. Although the concentrations of each butyltin compound varied greatly among different fish species, the concentrations of TBT and the total butyltin residues in the muscle tissues and liver of aquacultured cobia were much higher than those of the nonfarmed fishes caught from the coastal area of Taiwan. Similar to our results, Sasaki *et al.*[21] and Ueno *et al.*[22] also found that the concentrations of TBT or total butyltin compounds were much higher in aquacultured fishes than in wild fishes.

Similar to the situation in Taiwan, in Italy, four years after the restrictions on the usage of organotins, DBT was detected in the range of 1–26 ng/g and 1–4 ng/g wet weight, and TBT was detected in the range of 2–260 ng/g and 1–93 ng/g wet weight, respectively in both the farmed fishes and the free living fishes (including mussels)[23]. Six years after the ban on the use of TBT for all coastwise vessels and aquaculture facilities was enacted in Japan, the TBT concentration in the muscle of 11 species of fishes from the port of Osaka and Yodo River was detected to be in the range of 11–182 ng/g wet weight[19]. In the USA, six years after the controls on organotins were put into place in 1988, butyltin concentrations in fishes collected from the Gulf of Mexico during 1994 ranged between 158 and 289 ng/g wet weight. These results together with recent reports on the butyltin pollution in seawater and sediments[24,25,26] have shown that the sources of TBT contamination still remain worldwide, and that the partial ban on TBT usage was not sufficient to reduce the threat of butyltin compounds to human health.

III. *Status of Food Safety*

Seafood consumption is the main source of human dietary exposure to butyltin compounds. The toxicology of butyltin compounds in humans has not yet been fully resolved[27]. On the basis of TBT's ability to reduce the immune function, Penninks[28] suggested a tolerable daily intake (TDI) for TBT of 0.25 μg/kg body weight/day. Various attempts have thus been made to determine whether the level of organotin intake by humans from eating marine food should be a cause for concern.

The tolerable average residue level (TARL) is defined as the level in seafood that is tolerable for the average consumer with an average body weight of 60 kg.

TARL= (TDI × 60 kg body weight)/Average daily seafood consumption

In Taiwan an average 85.5 g/day/person of seafood was consumed in 1993–1996[29]. Using this value, an estimated 175.4 ng/g (wet weight) of TARL for TBT in seafood was obtained.

Penninks[28] found that DBT is equipotent or more toxic than TBT for mammals. He suggested that for the determination of toxicity both DBT and TBT should be considered. Heidrich *et al.*[30] also reported that human placental aromatase activity is directly inhibited by TBT (IC_{50} = 6.2 μM) or DBT but not by MBT and tetrabutyltin. Therefore, concentrations of DBT plus TBT in the muscle or the internal organ were taken while considering the residue level of butyltins.

Neither DBT nor TBT were detectable in 70% of the muscle and 87% of the internal organ of our samples. The highest concentrations of DBT plus TBT in the muscle and in the internal organs were found in *D. tumifrons* (103.8 ± 18.6 ng/g and 118.5 ± 81.1 ng/g, respectively) purchased in November. When the TBT concentration in seafood from markets in Asia, Australia, Europe, and North American were investigated, Keithly and his coworker[31] found that the TBT residues in marketable seafoods were consistent worldwide, averaging 185 ng/g dry weight. Considering the dry/wet ratio of fish muscle, TBT concentrations in seafood caught from the west coast of Taiwan were lower than those from other markets in the world. Concentration of DBT plus TBT in muscle and internal organs of all these tested fishes did not exceed the TARL value of 175.4 ng/g (wet weight). Based on our results, levels of DBT plus TBT observed in the muscle of fishes caught from the west coast of Taiwan were not matters of concern for human health.

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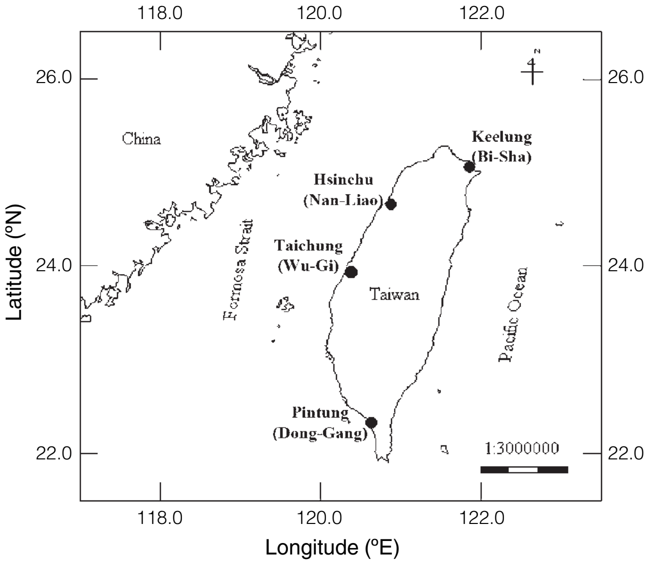

Figure 1. Map of sampling sites in Taiwan

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Table 1. Body length and body weight of the fishes bought from the fish markets

Date	Market location	Fishes	Body length (cm)*	Body weight (g)*
March 2006	Keelung	*Trichiurus lepturus*	82.5 (0.2)	765 (25)
		Psenopsis anomala	15.3 (0.8)	154 (9)
		Dentex tumifrons	15.3 (0.3)	168 (7)
		Polydactylus microstomus	27.0 (0.1)	370 (4)
	Hsinchu	*Trichiurus lepturus*	85.0 (0.2)	710 (10)
		Psenopsis anomala	14.1 (0.9)	104 (13)
		Dentex tumifrons	14.8 (0.3)	152 (8)
		Polydactylus microstomus	21.4 (0.7)	241 (7)
	Taichung	*Trichiurus lepturus*	60.0 (3.2)	281 (10)
		Acanthopagrus berda	18.0 (0.0)	221 (3)
		Nematalosa nasus	17.3 (1.5)	132 (8)
		Psettodes erumei	22.5 (0.7)	133 (14)
	Pingtung	*Trichiurus lepturus*	62.0 (1.3)	340 (5)
		Psenopsis anomala	13.9 (0.5)	100 (9)
		Acanthopagrus berda	22.3 (1.8)	326 (14)
		Polydactylus microstomus	24.4 (0.4)	241 (2)
June 2006	Keelung	*Trichiurus lepturus*	51.9 (2.8)	136 (23)
		Psenopsis anomala	15.7 (0.7)	157 (19)
		Dentex tumifrons	16.2 (6.4)	202 (6)
		Polydactylus microstomus	25.9 (0.1)	316 (11)
	Hsinchu	*Trichiurus lepturus*	60.0 (2.0)	228 (17)
		Psenopsis anomala	11.9 (0.5)	79 (8)
		Dentex tumifrons	12.3 (0.3)	85 (6)
		Polydactylus microstomus	21.8 (0.3)	201 (5)
	Taichung	*Trichiurus lepturus*	71.5 (16.3)	660 (57)
		Psenopsis anomala	12.0 (0.5)	78 (10)
		Psettodes erumei	21.5 (0.4)	167 (13)
		Polydactylus microstomus	21.8 (12.0)	220 (12)
	Pingtung	*Trichiurus lepturus*	50.5 (0.7)	163 (20)
		Psenopsis anomala	13.0 (0.7)	101 (13)
		Psettodes erumei	18.7 (0.3)	112 (13)
		Acanthopagrus berda	24.5 (3.4)	341 (12)
September 2006	Keelung	*Trichiurus lepturus*	54.0 (2.8)	169 (13)
		Psenopsis anomala	15.0 (0.7)	136 (18)
		Dentex tumifrons	16.3 (0.4)	182 (3)
		Polydactylus microstomus	25.5 (0.7)	293 (26)
		Babylonia formosae	5.5 (0.2)	17 (0)
		Lunella coronata	1.8 (0.1)	6 (0)
	Hsinchu	*Trichiurus lepturus*	50.8 (0.4)	160 (1)
		Psenopsis anomala	13.9 (0.1)	110 (2)
		Dentex tumifrons	14.0 (0)	106 (2)
		Polydactylus microstomus	22.0 (0.7)	206 (22)

Table 1. Continued

Date	Market location	Fishes	Body length (cm)*	Body weight (g)*
September 2006	Taichung	*Trichiurus lepturus*	48.5 (4.9)	148 (12)
		Pampus argenteus	12.8 (0.4)	94 (5)
		Dentex tumifrons	12.5 (0)	82 (9)
		Polydactylus microstomus	22.0 (1.4)	180 (13)
	Pingtung	*Trichiurus lepturus*	50.0 (2.8)	131 (16)
		Psenopsis anomala	13.8 (1.8)	114 (26)
		Dentex tumifrons	14.3 (1.1)	119 (23)
		Pennahia argentata	37.5 (3.5)	94 (6)
November 2006	Keelung	*Trichiurus lepturus*	67.5 (0.7)	292 (2)
		Psenopsis anomala	15.0 (0.6)	177 (1)
		Dentex tumifrons	15.0 (0.2)	144 (1)
		Polydactylus microstomus	26.0 (1.4)	330 (1)
		Babylonia formosae	5.0 (0.2)	16 (0)
		Lunella coronata	1.8 (0.1)	6 (0)
	Hsinchu	*Trichiurus lepturus*	67.5 (0.7)	312 (1)
		Psenopsis anomala	13.8 (0.4)	108 (3)
		Dentex tumifrons	13.6 (0.1)	104 (2)
		Polydactylus microstomus	22.0 (0.7)	272 (1)
	Taichung	*Trichiurus lepturus*	50.5 (0.7)	196 (3)
		Psenopsis anomala	13.9 (0.5)	119 (1)
		Pampus argenteus	14.6 (0.6)	198 (0)
		Polydactylus microstomus	20.5 (0.7)	218 (1)
	Pingtung	*Trichiurus lepturus*	58.5 (2.1)	206 (3)
		Psenopsis anomala	14.3 (0.4)	93 (4)
		Psettodes erumei	19.8 (1.1)	82 (14)
		Dentex tumifrons	14.3 (0.4)	138 (4)

Values are mean ± SD. Most samples are a pool of three to five organisms.
*Parentheses: standard deviation.

Table 2. Concentration of butyltin compounds (ng/g, wet weight), crude lipid content (%), and crude water content (%) in the dorsal muscle and the internal organs of *Trichiurus lepturus*

Date	Location	Tissue	MBT	DBT	TBT	TeBT	DBT+TBT	BTs*	Crude lipid content (%)	Crude water content (%)
March 2006	Keelung	Dorsal muscle	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	8.0 (0.2)	78.2
		Internal organs	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	11.4 (1.4)	52.9
	Hsinchu	Dorsal muscle	50.5 (15.1)	n.d.	n.d.	n.d.	n.d.	50.5 (15.1)	0.9 (0.3)	78.7
		Internal organs	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	3.7 (0)	60.9
	Taichung	Dorsal muscle	43.1 (24.9)	n.d	n.d.	n.d.	n.d.	43.1 (24.9)	0.2 (0.1)	80.8
		Internal organs	13.1 (1.8)	n.d.	n.d.	n.d.	n.d.	13.1 (1.8)	0.9 (0.1)	66.1
	Pingtung	Dorsal muscle	127.8 (2.6)	n.d.	n.d.	n.d.	n.d.	127.8 (2.6)	0.4 (0.2)	82.0
		Internal organs	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	3.3 (0)	49.1
June 2006	Keelung	Dorsal muscle	170.4 (28.3)	n.d.	n.d.	n.d.	n.d.	170.4 (28.3)	0.3 (0)	80.7
		Internal organs	290.7 (86.0)	n.d.	n.d.	n.d.	n.d.	290.7 (86.0)	1.3 (0)	78.9
	Hsinchu	Dorsal muscle	156.2 (15.5)	n.d.	n.d.	n.d.	n.d.	156.2 (15.5)	1.3 (0.1)	65.9
		Internal organs	102.0 (17.6)	n.d.	n.d.	n.d.	n.d.	102.0 (17.6)	3.7 (0)	51.1
	Taichung	Dorsal muscle	949.2 (49.1)	4.5 (0.6)	n.d.	n.d.	n.d.	953.7 (49.7)	1.0 (0.1)	55.0
		Internal organs	531.7 (3.1)	5.7 (1.6)	22.1 (3.9)	n.d.	27.8 (5.5)	559.5 (8.6)	2.4 (0)	80.5
	Pingtung	Dorsal muscle	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	0.4 (0.1)	68.8
		Internal organs	21.5 (3.4)	n.d.	n.d.	n.d.	n.d.	21.5 (3.4)	1.3 (0)	69.7
September 2006	Keelung	Dorsal muscle	133.7 (39.7)	n.d.	n.d.	n.d.	n.d.	133.7 (39.7)	1.9 (0.1)	80.5
		Internal organs	132.0 (79.2)	12.5 (5.7)	n.d.	n.d.	n.d.	144.5 (84.9)	2.1(0.1)	79.7
	Hsinchu	Dorsal muscle	178.7 (57.3)	26.8 (1.7)	n.d.	n.d.	26.8 (1.7)	205.5 (59.0)	0.6 (0.1)	85.2
		Internal organs	84.1 (5.6)	6.7 (0.8)	n.d.	n.d.	6.7 (0.8)	90.8 (6.4)	0.8 (0.1)	85.0
	Taichung	Dorsal muscle	62.4 (46.3)	56.2 (7.9)	n.d.	n.d.	56.2 (7.9)	118.6 (54.2)	1.3 (0.1)	80.3
		Internal organs	18.2 (4.8)	4.5 (0.2)	n.d.	n.d.	4.5 (0.2)	22.7 (5.0)	1.5 (0.1)	80.5
	Pingtung	Dorsal muscle	9.3 (4.9)	7.7 (1.9)	16.2 (9.9)	n.d.	23.9 (11.8)	33.2 (16.7)	0.2 (0)	79.9
		Internal organs	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	1.7 (0.1)	77.5
November 2006	Keelung	Dorsal muscle	85.8 (12.0)	4.8 (4.5)	6.4 (3.7)	n.d.	11.2 (8.2)	97.0 (20.2)	1.0 (0)	74.2
		Internal organs	80.0 (24.3)	21.2 (5.4)	38.9 (9.8)	n.d.	60.1 (15.2)	140.1 (39.5)	2.5 (0.1)	81.5
	Hsinchu	Dorsal muscle	19.2 (5.0)	n.d.	n.d.	n.d.	n.d.	19.2 (5.0)	0.9 (0)	76.1
		Internal organs	128.2 (94.2)	n.d.	n.d.	n.d.	n.d.	128.2 (94.2)	2.2 (0.1)	83.6
	Taichung	Dorsal muscle	101.6 (35.0)	24.0 (16.5)	4.7 (5.1)	n.d.	28.7 (21.6)	130.3 (56.6)	1.4 (0.1)	80.0
		Internal organs	42.1 (11.8)	n.d.	n.d.	n.d.	n.d.	42.1 (11.8)	2.1 (0)	72.1
	Pingtung	Dorsal muscle	73.9 (43.3)	16.4 (9.5)	n.d.	n.d.	16.4 (9.5)	90.3 (52.8)	0.3 (0)	76.9
		Internal organs	210.8 (93.3)	13.0 (75.1)	12.3 (9.5)	n.d.	25.3 (84.6)	236.1 (177.9)	1.8 (0.1)	78.7

Values are mean ± SD. Most samples are a pool of three to five organisms.
*BTs = MBT + DBT + TBT + TeBT. Parentheses: standard deviation. n.d.: not detected.

Table 3. Concentration of butyltin compounds (ng/g, wet weight), crude lipid content (%), and crude water content (%) in the dorsal muscle and the internal organs of *Psenopsis anomala*.

Date	Location	Tissue	MBT	DBT	TBT	TeBT	DBT+TBT	BTs*	Crude lipid content (%)	Crude water content (%)
March 2006	Keelung	Dorsal muscle	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	1.5 (0.4)	79.6
		Internal organs	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	14.4 (0)	50.7
	Hsinchu	Dorsal muscle	34.2 (4.8)	24.9 (10.6)	n.d.	n.d.	24.9 (10.6)	59.1 (15.4)	0.9 (0.3)	72.5
		Internal organs	21.8 (2.0)	n.d.	n.d.	n.d.	n.d.	21.8 (2.0)	3.7 (0)	59.9
	Pingtung	Dorsal muscle	327.0 (19.5)	n.d.	36.0 (5.1)	n.d.	36.0 (5.1)	363.0 (24.6)	3.9 (0.2)	80.0
		Internal organs	247.2 (11.9)	n.d.	n.d.	n.d.	n.d.	247.2 (11.9)	8.1 (2.8)	58.1
June 2006	Keelung	Dorsal muscle	799.5 (4.8)	n.d.	n.d.	n.d.	n.d.	799.5 (4.8)	5.0 (0.1)	77.1
		Internal organs	555.3 (16.9)	n.d.	n.d.	n.d.	n.d.	555.3 (16.9)	7.6 (0.1)	87.7
	Hsinchu	Dorsal muscle	853.4 (45.0)	n.d.	13.2 (3.4)	n.d.	13.2 (3.4)	866.6 (48.4)	1.0 (0)	65.8
		Internal organs	286.1 (36.5)	n.d.	n.d.	n.d.	n.d.	286.1 (36.5)	3.6 (0)	57.5
	Pingtung	Dorsal muscle	3.6 (0.4)	n.d.	n.d.	n.d.	n.d.	3.6 (0.4)	0.5 (0)	82.7
		Internal organs	627.8 (74.4)	5.0 (1.6)	n.d.	n.d.	5.0 (1.6)	632.8 (76.0)	28.0 (0.1)	90.5
September 2006	Keelung	Dorsal muscle	257.7 (31.3)	4.6 (3.8)	7.4 (1.0)	n.d.	12.0 (4.8)	269.7 (36.1)	7.2(0.6)	73.6
		Internal organs	22.2 (1.9)	4.9 (0.6)	n.d.	n.d.	4.9 (0.6)	27.1 (2.5)	13.2(0.4)	71.0
	Hsinchu	Dorsal muscle	152.4 (32.2)	25.3 (1.6)	n.d.	n.d.	25.3 (1.6)	177.7 (33.8)	5.1 (0.3)	74.1
		Internal organs	53.1 (3.3)	8.1 (3.5)	n.d.	n.d.	8.1 (3.5)	61.2 (6.8)	7.6 (0.3)	64.2
	Pingtung	Dorsal muscle	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	0.5 (0)	80.0
		Internal organs	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	1.2 (0.1)	79.9
November 2006	Keelung	Dorsal muscle	155.6 (31.6)	n.d.	n.d.	n.d.	n.d.	155.6 (31.6)	3.1 (0.5)	76.3
		Internal organs	105.7 (44.7)	30.6 (8.7)	38.6 (2.9)	n.d.	69.2 (11.6)	174.9 (56.3)	4.3 (0.1)	79.2
	Hsinchu	Dorsal muscle	84.1 (42.4)	11.1 (26.6)	12.3 (26.6)	n.d.	23.4 (53.2)	107.5 (95.6)	3.5 (0.1)	76.2
		Internal organs	51.7 (29.3)	n.d.	n.d.	n.d.	n.d.	51.7 (29.3)	5.3 (0.1)	74.4
	Taichung	Dorsal muscle	108.9 (46.8)	24.7 (7.9)	12.7 (44.3)	n.d.	37.4 (52.2)	146.3 (99.0)	5.5 (0.2)	79.7
		Internal organs	73.7 (12.7)	n.d.	n.d.	n.d.	n.d.	73.7 (12.7)	11.1 (0.2)	78.6
	Pingtung	Dorsal muscle	110.1 (26.5)	30.5 (13.1)	10.4 (10.3)	n.d.	40.9 (23.4)	151.0 (49.9)	3.1 (0.1)	74.0
		Internal organs	50.2 (26.2)	6.9 (98.1)	n.d.	n.d.	6.9 (98.1)	57.1 (111.4)	3.5 (0.1)	75.2

Values are mean ± SD. Most samples are a pool of three to five organisms.
*BTs = MBT + DBT + TBT + TeBT. Parentheses: standard deviation. n.d.: not detected.

Table 4. Concentration of butyltin compounds (ng/g, wet weight), crude lipid content (%), and crude water content (%) in the dorsal muscle and the internal organs of *Dentex tumifrons*

Date	Location	Tissue	MBT	DBT	TBT	TeBT	DBT+TBT	BTs*	Crude lipid content (%)	Crude water content (%)
March 2006	Keelung	Dorsal muscle	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	0.4 (0.2)	80.1
		Internal organs	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	2.9 (0.4)	51.6
	Hsinchu	Dorsal muscle	122.5 (16.0)	n.d.	70.8 (10.0)	n.d.	70.8 (10.0)	193.3 (26.0)	5.3 (0.3)	78.6
		Internal organs	6.5 (0.9)	n.d.	n.d.	n.d.	n.d.	6.5 (0.9)	22.7 (3.1)	53.3
June 2006	Keelung	Dorsal muscle	220.5 (84.7)	n.d.	n.d.	n.d.	n.d.	220.5 (84.7)	0.2 (0)	65.9
		Internal organs	134.0 (70.6)	n.d.	n.d.	n.d.	n.d.	134.0 (70.6)	2.0 (0.1)	64.0
	Hsinchu	Dorsal muscle	84.6 (3.0)	n.d.	n.d.	n.d.	n.d.	84.6 (3.0)	0.4 (0)	67.8
		Internal organs	127.4 (14.7)	4.2 (0.6)	n.d.	n.d.	4.2 (0.6)	131.6 (15.3)	3.6 (0.1)	56.8
September 2006	Keelung	Dorsal muscle	171.3 (12.0)	26.3 (8.2)	n.d.	n.d.	26.3 (8.2)	197.6 (20.2)	2.2(0.1)	77.7
		Internal organs	65.1 (23.9)	14.7 (4.2)	n.d.	103.8 (76.9)	118.5 (81.1)	183.6 (105.0)	12.5(0.1)	77.6
	Hsinchu	Dorsal muscle	56.7 (0.1)	30.6 (9.0)	n.d.	n.d.	30.6 (9.0)	87.3 (9.1)	0.3 (0)	77.0
		Internal organs	114.7 (11.0)	35.2 (11.7)	n.d.	n.d.	35.2 (11.7)	149.9 (22.7)	4.1 (0.1)	74.4
	Taichung	Dorsal muscle	78.6 (27.6)	17.1 (3.8)	n.d.	n.d.	17.1 (3.8)	95.7 (31.4)	5.3 (0.2)	78.6
		Internal organs	28.2 (7.5)	8.1 (1.4)	n.d.	n.d.	8.1 (1.4)	36.3 (8.9)	10.1 (0.2)	71.7
	Pingtung	Dorsal muscle	n.d.	n.d.	8.6 (0.8)	n.d.	8.6 (0.8)	8.6 (0.8)	3.5 (0.1)	77.8
		Internal organs	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	5.2 (0)	80.0
November 2006	Keelung	Dorsal muscle	45.5 (9.1)	n.d.	37.2 (4.3)	n.d.	37.2 (4.3)	82.7 (13.4)	1.4 (0)	79.1
		Internal organs	100.8 (25.3)	26.7 (7.8)	23.4 (4.8)	n.d.	50.1 (12.6)	150.9 (37.9)	3.2 (0.1)	66.8
	Hsinchu	Dorsal muscle	158.2 (49.3)	21.5 (6.3)	n.d.	7.2 (7.4)	21.5 (6.3)	186.9 (63.0)	1.1 (0)	79.5
		Internal organs	33.6 (13.5)	n.d.	n.d.	n.d.	n.d.	33.6 (13.5)	3.2 (0.1)	71.2
	Pingtung	Dorsal muscle	136.7 (26.0)	48.2 (9.3)	55.6 (9.3)	n.d.	103.8 (18.6)	240.5 (44.6)	3.7 (0.1)	78.5
		Internal organs	80.4 (29.3)	5.9 (6.1)	8.7 (7.5)	n.d.	14.6 (13.6)	95.0 (42.9)	6.1 (0)	72.0

Values are mean ± SD. Most samples are a pool of three to five organisms.
*BTs= MBT + DBT + TBT + TeBT. Parentheses: standard deviation. n.d.: not detected.

Table 5. Concentration of butyltin compounds (ng/g, wet weight), crude lipid content (%), and crude water content (%) in the dorsal muscle and the internal organs of *Polydactylus microstomus*

Date	Location	Tissue	MBT	DBT	TBT	TeBT	DBT+TBT	BTs*	Crude lipid content (%)	Crude water content (%)
March 2006	Keelung	Dorsal muscle	n.d.	n.d.	n.d.	15.0 (3.4)	n.d.	15.0 (3.4)	10.2 (2.7)	63.8
		Internal organs	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	21.7 (2.2)	53.2
	Hsinchu	Dorsal muscle	3.7 (1.8)	n.d.	20.4 (2.9)	n.d.	20.4 (2.9)	24.1 (4.7)	3.9 (0.3)	72.6
		Internal organs	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	37.6 (2.4)	51.8
	Pingtung	Dorsal muscle	262.2 (76.4)	n.d.	n.d.	n.d.	n.d.	262.2 (76.4)	3.6 (0.1)	70.8
		Internal organs	295.5 (6.4)	n.d.	6.8 (0.9)	n.d.	6.8 (0.9)	302.3 (7.3)	21.1 (2.8)	55.7
June 2006	Keelung	Dorsal muscle	262.7 (37.6)	n.d.	n.d.	n.d	n.d.	262.7 (37.6)	5.5 (0.1)	55.6
		Internal organs	596.5 (19.0)	n.d.	n.d.	n.d.	n.d.	596.5 (19.0)	22.6 (0.2)	61.3
	Hsinchu	Dorsal muscle	386.4 (25.1)	n.d.	n.d.	n.d.	n.d.	386.4 (25.1)	7.1 (0.8)	58.0
		Internal organs	44.3 (10.0)	n.d.	n.d.	n.d.	n.d.	44.3 (10.0)	32.6 (0)	45.7
	Taichung	Dorsal muscle	554.3 (17.8)	n.d.	n.d.	n.d.	n.d.	554.3 (17.8)	11.9 (0)	64.8
		Internal organs	63.7 (32.7)	n.d.	n.d.	n.d.	n.d.	63.7 (32.7)	20.5 (0.6)	54.0
September 2006	Keelung	Dorsal muscle	482.6 (128.7)	6.0 (1.9)	n.d.	n.d.	6.0 (1.9)	488.6 (130.6)	2.8(0.2)	78.8
		Internal organs	41.8 (19.0)	6.0 (0.6)	n.d.	n.d.	6.0 (0.6)	47.8 (19.6)	4.8(0.1)	77.7
	Hsinchu	Dorsal muscle	110.7 (9.1)	27.6 (2.6)	n.d.	n.d.	27.6 (2.6)	138.3 (11.7)	4.1 (0.5)	81.9
		Internal organs	170.2 (18.9)	11.8 (0.6)	6.7 (1.0)	n.d.	18.5 (1.6)	188.7 (20.5)	7.6 (0.3)	80.9
	Taichung	Dorsal muscle	63.5 (24.7)	n.d.	n.d.	n.d.	n.d.	63.5 (24.7)	1.3 (0.1)	81.2
		Internal organs	8.0 (0.1)	n.d.	n.d.	n.d.	n.d.	8.0 (0.1)	3.7 (0.1)	86.7
November 2006	Keelung	Dorsal muscle	95.3 (30.1)	n.d.	n.d.	n.d.	n.d.	95.3 (30.1)	2.9 (0.1)	70.4
		Internal organs	41.1 (18.9)	17.8 (3.5)	n.d.	n.d.	17.8 (3.5)	58.9 (22.4)	4.6 (0.6)	68.8
	Hsinchu	Dorsal muscle	116.2 (14.7)	n.d.	n.d.	n.d.	n.d.	116.2 (14.7)	7.9 (0.1)	66.0
		Internal organs	41.5 (10.0)	4.7 (4.6)	n.d.	n.d.	4.7 (4.6)	46.2 (14.6)	8.6 (0.6)	52.8
	Taichung	Dorsal muscle	72.4 (30.2)	n.d.	n.d.	n.d.	n.d.	72.4 (30.2)	1.5 (0.1)	66.6
		Internal organs	43.4 (15.6)	n.d.	n.d.	n.d.	n.d.	43.4 (15.6)	4.1 (0.1)	52.5

Values are mean ± SD. Most samples are a pool of three to five organisms.
*BTs = MBT + DBT + TBT + TeBT. Parentheses: standard deviation. n.d.: not detected.

Table 6. Concentration of butyltin compounds (ng/g, wet weight), crude lipid content (%), and crude water content (%) in the dorsal muscle and the internal organs of *Acanthopagrus berda*, *Nematalosa nasus*, *Psettodes erumei*, *Pampus argenteus*, *Pennahia argentata*, *Babylonia formosae*, and *Lanella coronata*

Date	Location	Tissue	MBT	DBT	TBT	TeBT	DBT+TBT	BTs*	Crude lipid content (%)	Crude water content (%)
					Acanthopagrus berda					
March 2006	Taichung	Dorsal muscle	326.7 (35.5)	n.d.	n.d.	n.d.	n.d.	326.7 (35.5)	0.7 (0.5)	77.0
		Internal organs	112.1 (23.2)	n.d.	10.5 (1.5)	n.d.	10.5 (1.5)	122.6 (24.7)	2.0 (0.1)	50.3
	Pingtung	Dorsal muscle	137.4 (51.7)	n.d.	n.d.	n.d.	n.d.	137.4 (51.7)	1.0 (0.4)	76.7
		Internal organs	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	2.2 (0.2)	48.3
June 2006	Pingtung	Dorsal muscle	549.0 (21.9)	n.d.	n.d.	25.4 (3.0)	n.d.	574.4 (24.9)	0.8 (0)	75.6
		Internal organs	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	2.1 (0)	50.2
					Nematalosa nasus					
March 2006	Taichung	Dorsal muscle	385.5 (85.3)	n.d.	n.d.	n.d.	n.d.	385.5 (85.3)	0.1 (0)	72.7
		Internal organs	307.2 (80.6)	n.d.	6.1 (0.6)	n.d.	6.1 (0.6)	313.3 (81.2)	5.3 (0.5)	55.3
					Psettodes erumei					
March 2006	Taichung	Dorsal muscle	82.7 (47.0)	n.d.	n.d.	n.d.	n.d.	82.7 (47.0)	1.0 (0)	78.3
		Internal organs	112.1 (23.2)	n.d.	n.d.	n.d.	n.d.	112.1 (23.2)	1.6 (0.1)	68.7
June 2006	Taichung	Dorsal muscle	95.8 (9.6)	n.d.	n.d.	n.d.	n.d.	95.8 (9.6)	1.1 (0.1)	77.5
		Internal organs	518.0 (25.0)	n.d.	n.d.	n.d.	n.d.	518.0 (25.0)	1.9 (0.1)	66.3
	Pingtung	Dorsal muscle	447.1 (30.2)	7.3 (2.0)	n.d.	n.d.	7.3 (2.0)	454.4 (32.2)	0.4 (0.1)	84.3
		Internal organs	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	31.9 (0.1)	78.7
November 2006	Pingtung	Dorsal muscle	80.7[a]	23.5[a]	14.0[a]	n.d.	37.5[a]	118.2[a]	10.3 (0.5)	80.0
		Internal organs	102.1[a]	n.d.	n.d.	n.d.	n.d.	102.1[a]	38.9 (0.7)	76.1
					Pampus argenteus					
September 2006	Taichung	Dorsal muscle	42.2 (0.1)	34.2 (0.4)	n.d.	n.d.	34.2 (0.4)	76.4 (0.5)	1.4 (0)	74.1
		Internal organs	6.6 (0.9)	n.d.	n.d.	n.d.	n.d.	6.6 (0.9)	3.3 (0.1)	71.4
November 2006	Taichung	Dorsal muscle	84.3 (18.8)	13.9 (15.6)	n.d.	n.d.	13.9 (15.6)	98.2 (34.4)	1.1 (0.1)	75.5
		Internal organs	134.8 (21.3)	4.8 (8.6)	n.d.	n.d.	4.8 (8.6)	139.6 (29.9)	1.3 (0.1)	89.2
					Pennahia argentata					
September 2006	Pingtung	Dorsal muscle	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	0.7 (0)	72.7
		Internal organs	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	10.9 (0.3)	55.3

Table 6. Continued

Date	Location	Tissue	MBT	DBT	TBT	TeBT	DBT+TBT	BTs*	Crude lipid content (%)	Crude water content (%)
					Babylonia formosae					
September 2006	Keelung	Dorsal muscle	48.0 (6.8)	n.d.	n.d.	n.d.	n.d.	48.0 (6.8)	12.9(0.1)	72.3
November 2006	Keelung	Dorsal muscle	22.9[a]	n.d.	n.d.	n.d.	n.d.	22.9[a]	7.5 (0.2)	72.6
					Lanella coronata					
September 2006	Keelung	Dorsal muscle	141.7 (10.3)	n.d.	n.d.	n.d.	n.d.	141.7 (10.3)	13.7(0.3)	77.1
November 2006	Keelung	Dorsal muscle	148.3 (4.3)	34.9 (20.9)	11.3 (19.6)	n.d.	46.2 (40.5)	194.5 (44.8)	6.2 (0.1)	74.2

Values are mean ± SD. Most samples are a pool of three to five organisms.
[a]Only one organism is used for the test.
*BTs = MBT + DBT + TBT + TeBT. Parentheses: standard deviation. n.d.: not detected.