Characteristics of Marine Litters Distribution on the Seabed of South Sea and West Sea of Korea, and East China Sea

ToxEHS

Organotin Compounds in Olive Flounder *Paralichthys Olivaceus* and Flathead Mullet *Mugil cephalus* from Korean Coastal Waters

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Abstract

Concentrations of organotin compounds (OTCs) were measured in the muscle and liver of two fish species (*Paralichthys Olivaceus* and *Mugil cephalus*) collected in the six developed coastal regions of Korea: Incheon, Gunsan, Yeosu, Masan, Pohang and Jeju. Concentrations of butyltin compounds ($\sum$BTs) in the muscle and liver of the two fish species were detected in the range of 1.7-28.8 (mean: 8.8) ng/g-wet wt. as Cl and 18.7-5,096.9 (mean: 832.6) ng/g-wet wt. as Cl, respectively. Concentrations of phenyltin compounds ($\sum$PTs) ranged <0.1-72.5 (mean: 10.89) ng/g-wet wt. as Cl in the muscle and <0.1-316.3 (mean: 28.0) ng/g-wet wt. as Cl in the liver. Concentrations of OTCs in the liver were higher than those of OTCs in the muscle. The compositions of $\sum$BTs in the muscle and liver were different from each other, but those of $\sum$PTs were similar. Tributytin (TBT) and triphenytin (TPT) levels in the two fish species collected from Korean coastal waters were significantly lower than the tolerable average residue level (TARL) estimated based on the tolerable daily intake (TDI).

Keywords: Organotin compounds, Coastal fish, Tolerable average residue levels (TARL)

Introduction

Organotin compounds (OTCs) were initially synthesized in the early part of the 1850s and the amount of their commercial use has rapidly increased since then[1]. OTCs have been used mainly as an anti-fouling paint for the bottom of a ship or as antifouling agent for fishing net, and the demand for them has increased because they help reduce the costs of operating and maintaining a ship with their cost-benefits of high anti-fouling effects against adhesive organisms like barnacle[2]. However, the use of OTCs is restricted or even banned in many countries[3], since it was reported in the 1970s that they are substances that contribute to abnormality in oyster shells[4,5] and imposex (a genital disruption phenomenon) in gastropods[6-8], and it was also revealed that they, even in low concentration, have a high toxicity to living organisms and will persist in higher animals[9]. In Korea, form March 2003, the use of TBT as an anti-fouling paint for ships has been regulated and totally banned[10]. However, ever since the ban was put in place, they have been continually detected in various matters like seawater, marine deposits and marine life from coastal region waters in relatively high concentrations; therefore, the need for a continuous monitoring has been discussed[11,12].

This research attempted to investigate the body residue concentration and enrichment characteristics of OTCs detected in the two species of coastal fish that are useful fisheries resources of the marine products that account for supplying about 40% of the protein sources for the Korean people. Additionally, by utilizing national statistical data, the amount of OTCs taken from the two fish species was evaluated. These results will be used as basic data to evaluate the contamination levels of organotin compounds in marine life immediately after the regulations were put in place.

Results and Discussion

Body Residue and Bio-concentration Characteristics

Table 2 and Table 3 show the concentrations of OTCs

Table 1. Analytical conditions of GC-MS for Organotin compounds.

	Analytical conditions	
GC	column career gas injection port temp. injection mode: column flow: oven temp. programming	J&W DB-5 (30 m × 0.25 mm × 0.25 μm) He (99.999%) 250°C splitless, purge time: 1 min, 2 μL 1.0 mL/min 10°C/min 4°C/min 10°C/min 70°C → 120°C → 250°C → 280°C
MS	interface temp ionization mode: electron energy: ion source temp: analyzer: detection mode:	280°C electron impact (EI) 70 eV 280°C quardrupole Selected Ion Monitoring (SIM)

Table 2. Concentrations of organotin compounds in muscle and liver of olive flounder, *Paralichthys Olivaceus*.

		West sea		South sea		East sea	Jeju		
		Incheon	Gunsan	Masan	Yeosu	Pohang	Jeju	Range	Mean
		Conc. (ng/g-wet wt. as Cl)							
MBT	Muscle	<0.1	<0.1	<0.1	<0.1	<0.1	<0.1	–	–
	Liver	<0.1	10.2	200.2	245.5	461.2	4.5	<0.1-461.2	153.6
DBT	Muscle	<0.1	0.9	4.4	<0.1	7.6	<0.1	<0.1-7.6	2.2
	Liver	754.0	64.8	113	1,126.2	3,932.7	42.4	42.4-3,932.7	1,005.5
TBT	Muscle	3.5	5.05	17.1	9.4	21.2	2.0	2.0-21.2	9.7
	Liver	29.8	31.4	42.4	429.4	703.1	13.1	13.1-703.1	208.2
∑BTs	Muscle	3.5	6.0	21.5	9.4	28.7	2.0	2.0-28.8	11.9
	Liver	783.7	106.0	355.6	1,801.2	5,096.9	60.0	60.0-5,096.9	1,367.2
MPT	Muscle	<0.1	<0.1	1.6	<0.1	<0.1	<0.1	<0.1-1.6	0.3
	Liver	<0.1	<0.1	<0.1	<0.1	<0.1	<0.1	–	–
DPT	Muscle	<0.1	<0.1	0.56	0.2	0.11	<0.1	<0.1-0.6	0.1
	Liver	<0.1	<0.1	<0.1	<0.1	<0.1	<0.1	–	–
TPT	Muscle	<0.1	2.8	70.3	6.9	33.1	11.9	<0.1-70.3	20.8
	Liver	<0.1	<0.1	<0.1	<0.1	316.3	<0.1	<0.1-316.3	52.7
∑PTs	Muscle	<0.1	2.8	72.5	7.1	33.2	12.0	<0.1-72.5	21.3
	Liver	<0.1	<0.1	<0.1	<0.1	316.3	<0.1	<0.1-316.3	52.7

in the body of olive flounder and flathead mullet collected from each of the regions. The concentrations of butyltin compounds (∑BTs: MBT+DBT+TBT) detected in the muscle and liver of olive flounder ranged 2.0-28.8 (mean: 11.9) ng/g-wet wt. as Cl and 60.0-5,096.9 (mean: 1,367.2) ng/g-wet wt. as Cl, respectively; the concentrations of phenyltin compounds (∑PTs: MPT+DPT+TPT) ranged <0.1-72.5 (mean: 21.6) ng/g-wet wt. as Cl and <0.1-316.3 (mean: 52.7) ng/g-wet wt. as Cl, respectively. The concentrations of ∑BTs detected in the muscle and liver of flathead mullet ranged 1.7-15.5 (mean: 5.7 ng/g-wet wt. as Cl and 18.7-1,181.0 (mean: 297.9) ng/g-wet wt. as Cl, respectively; the concentrations of ∑PTs ranged <0.1-2.0 (mean: 0.5) ng/g-wet wt. as Cl and <0.1-19.8 (mean: 3.30) ng/g-wet wt. as Cl, respectively. The concentrations of the two compounds were relatively higher in the liver than in the muscle, which is an edible portion. These results are similar to those obtained by other researchers and they imply that OTCs, a lipophilic substance, are detected in the liver in higher concentrations since the fat content in the liver is more than the fat content in the muscle[13-15].

The concentration of OTCs in the body of flathead mullet was relatively lower than that of olive flounder. The concentration of OTCs in an organism takes two forms: indirect concentration from feeds and direct concentration from water environment[16]. Indirect bio-

Table 3. Concentrations of organotin compounds in muscle and liver of flathead mullet, *Mugil cephalus*.

		West sea		South sea		East sea	Jeju	Range	Mean
		Incheon	Gunsan	Masan	Yeosu	Pohang	Jeju		
		Conc. (ng/g-wet wt. as Cl)							
MBT	Muscle	<0.1	<0.1	0.2	<0.1	0.5	0.7	<0.1-0.7	0.2
	Liver	97.1	41.8	2.8	122.0	85.4	51.3	2.8-122.0	66.7
DBT	Muscle	3.9	<0.1	0.6	<0.1	1.0	0.8	<0.1-3.9	1.0
	Liver	107.7	17.7	1.5	443.1	18.6	20.0	1.5-443.1	101.4
TBT	Muscle	1.6	1.7	14.7	3.0	1.1	4.8	1.1-14.7	4.5
	Liver	121.2	12.2	14.5	615.9	7.9	6.8	6.8-615.9	129.7
∑BTs	Muscle	5.5	1.7	15.5	3.0	2.5	6.2	1.7-15.5	5.7
	Liver	326.0	71.7	18.7	1,181.0	111.9	78.1	18.7-1,181.0	297.9
MPT	Muscle	<0.1	<0.1	<0.1	<0.1	<0.1	<0.1	–	–
	Liver	<0.1	<0.1	<0.1	<0.1	<0.1	<0.1	–	–
DPT	Muscle	<0.1	<0.1	<0.1	<0.1	<0.1	<0.1	–	–
	Liver	<0.1	<0.1	<0.1	<0.1	<0.1	<0.1	–	–
TPT	Muscle	<0.1	<0.1	2.0	<0.1	0.3	0.7	<0.1-2.0	0.5
	Liver	<0.1	<0.1	19.8	<0.1	<0.1	<0.1	<0.1-19.8	3.3
∑PTs	Muscle	<0.1	<0.1	2.0	<0.1	0.3	0.7	<0.1-2.0	0.5
	Liver	<0.1	<0.1	19.8	<0.1	<0.1	<0.1	<0.1-19.8	3.3

concentration is associated with various causes like the size, age and feeding habit of organisms[17], while direct concentration is decided by the elution of OTCs from a ship and fishing net and/or the elution of OTCs accumulated in sediments. In particular, the concentration of OTCs is higher in marine sediments than in seawater[18], and the flows of seawater may gush forth into the water in which the OTCs are accumulated in marine sediments in relatively higher concentration[19]. Olive flounder is a benthic fish which preys on bottom -dwelling polychaetes and inhabits on marine sediments with relatively higher OTCs concentrations compared to seawater. In contrast, flathead mullet is an omnivorous fish that usually inhabits in the surface layer of waters and exposed to marine sediments only when to catch benthic preys. It is deemed that the difference in the concentration of OTCs accumulated in the two fish species is due to the differences in the feeding levels and the environment of their habitats.

In terms of regional variation, the concentration of ∑BTs detected in the muscle of olive flounder was highest at 28.8 ng/g-wet wt. as Cl in Pohang, located on the east coast, while that of ∑PTs was highest at 70.2 ng/g-wet wt. as Cl in Masan on the south coast. As for flathead mullet, the concentration of ∑BTs and ∑PTs marked highest at 15.5 ng/g-wet wt. as Cl and 2.0 ng/g-wet wt. as Cl, respectively, in Masan. These regions are comprised of ports and dockyards which are known to be major contamination sources[20]. National major industrial facilities are also located in the

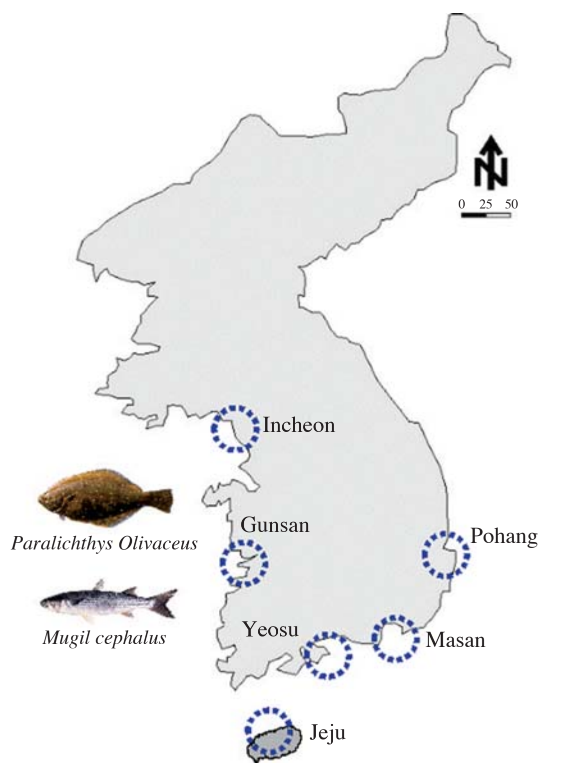

Figure 1. Sampling locations and fish species for analysis.

regions with other harmful chemicals being detected in relatively higher concentrations[21,22]. It appears that the

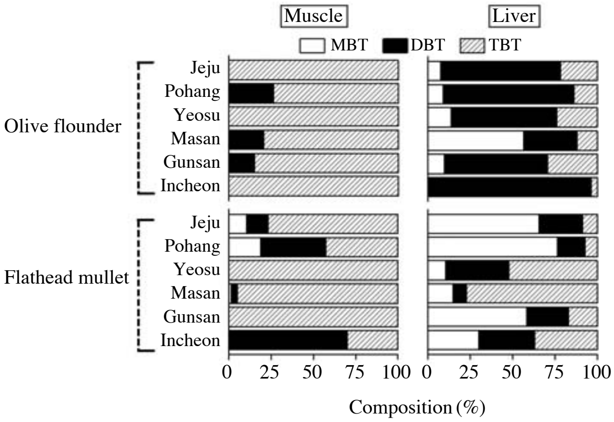

Figure 2. Composition of butyltin compounds to the total butyltin concentrations in the muscle and liver of two fish species.

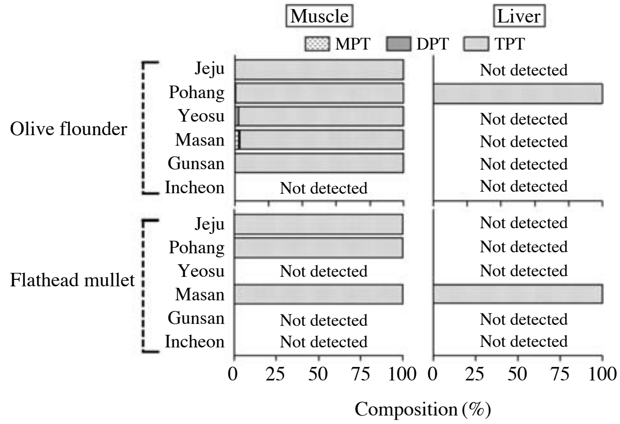

Figure 3. Composition of phenyltin compounds to the total phenyltin concentrations in the muscle and liver of two fish species.

results of this research also demonstrate these facts.

Figures 2 and 3 show the composition of $\sum$BTs and $\sum$PTs detected in the body of olive flounder and flathead mullet. The compositions of TBT, from among $\sum$BTs, detected in the two fish species were $81.8 \pm 23.7\%$ for the muscle and $25.4 \pm 21.5\%$ for the liver, whereby implying that the compositions of DBT and MBT, which are metabolites, are high in the liver. This is possibly due to the metal binding proteins like glutathione exist in relatively smaller quantities in the muscle than in the liver, which has detoxifying properties and in which metabolism is actively conducted[14,15]. Meanwhile, DBT and MBT are limitedly used as wood preservers for wood, PVC stabilizer, or industrial catalyte[23]. The results of this research imply that these substances were not directly accumulated in the body but metabolites of TBT. As for TPT, from among $\sum$PTs, the compositions of metabolized products MPT and DPT were low both in the muscle and the liver. Harino and others (2000)[24] reported that the composition of TPT detected from Japanese sea perch, white croaker, and yellowtail is higher than that of TBT because TPT is decomposed at a lower speed than TBT. The results of this research also demonstrate that TPT is relatively hard to decompose.

The concentration of $\sum$BTs in the body of the two fish species discovered in this research was compared to the results of other researches and it was found that the concentration showed a similar level in all the regions except in Osaka Bay in Japan (Table 4). The concentration range detected in fishes caught in Osaka Bay was relatively high, but Harino (2000)[24] reported that it was not high enough to have ill effects on the health of a human body. Nevertheless, continuous monitoring are important and need be conducted since OTCs are still detected from creatures and water environment even after the use of the substance is banned[11,12] and Koreans consume marine products in relatively greater quantities.

Evaluating the Intake

Penninks (1993)[25] suggested that the tolerable daily intake (TDI) of TBT should be 0.25 μg/kg body weight /day and the World Health Organization (WHO) set a provisional TDI for TPT at 0.50 μg/kg/day[26]. Considering that the average weight of Koreans is 55 kg[27], TDI for Koreans is 13.8 μg/day for TBT and 27.5 μg/day for TPT. Here, when TDI is divided by the average daily intake for olive flounder and flathead mullet, tolerable average residue levels for intake can be calculated. Since the daily average intake for olive flounder is 1.5 g and for flathead mullet is less than 0.1 g[28], it is considered that the exposure of organotin compounds caused by the intake of the two fish species is considerably small. In addition, since the frequency of fish intake for Koreans is less than once a week, it is determined that the actual amount of TBT and TPT taken would be even smaller.

Conclusions

Analyzing OTCs contained in olive flounder and flathead mullet inhabiting Korean coasts, it was found that OTCs detected in olive flounder were relatively higher in concentration than those detected in flathead mullet; in terms of body portions, OTCs in the muscle, an edible portion, were lower in concentration than those in the liver, which neutralizes poison. It seemed that, compared to flathead mullet, olive flounder, which

Table 4. Comparison of total butyltin concentrations in fish muscle of this study with concentrations reported from other studies.

Countries	Location	Species	SBT	Unit	Refs.
Australia	Sydney, Brisbane, Perth, Tasmania, Townsville, Atherton	Rubberlip morwong, Shovelnose ray, Blue groper, Long-spined snapper, Sea mullet, Spiny tailed leatherjacket, Australian herring, Striped seaperch, Sea mullet, Black bream, Rainbow trout, Atlantic salmon, Mud flathead, Silver bream, Silver trevally Stripey, Black pomfret, Squid, Seabass	ND*-47	ng/g wet-wt.	
Papua New Guinea	Port Moresby	Sea mullet, Mud clam, Tilapia, Crab	ND*-9.0	ng/g wet-wt.	
Solomon Islands	Honiara	Greenspotted kingfish, Indian mackerel, Paddletail snapper	0.2-1.4	ng/g wet-wt.	
India	Delhi, Porto Novo, Bombay, Calcutta	Scombrid, Catfish, Jawfish, Sciaenid fish, Indian mackerel, Prawn, Sea mullet, Pearl spot, Catla, Silver pomfret, Black bream, Threadfins, Perch	ND*-79	ng/g wet-wt.	Kannan *et al.* [1995][14]
Bangladesh	Dhaka	Flounder, Catla, Hilsa, Aor	0.47-190	ng/g wet-wt.	
Thailand	Bangkok	Silver pomfret, Indian mackerel, Giant seaperch	2.9-16	ng/g wet-wt.	
Vietnam	Hanoi, Phu Da, Ho Chi Minh, Cu Chi	Silver carp, Carp, Bream, Sea mullet, Perch, Sea mullet, Tilapia, Prawn, Shellfish	ND*-1.7	ng/g wet-wt.	
Indonesia	Bogor	Big-eyed scad, Deep-bodied crucian carp	0.41-19	ng/g wet-wt.	
Taiwan	Taipei	Tilapia, Milkfish, Seabream, Bivalve	0.49-18	ng/g wet-wt.	
Taiwan[1)]	Kaohsiung	*Pagrus major, Cephalopholis argus, Lates calacriter, Scarus cedema*	18.5-54.6	ng/g dry-wt	Hung *et al.* [1998][31]
Japan	Osaka bay	Japanese sea perch, White croaker, Yellowtail, Bandfish, Silver whiting, File fish, Triped puffer, Grey mullet, Japanese barbel, Bluegill	57-251	ng/g wet-wt.	Harino [2000][24]
Korea	Incheon, Gunsan, Masan, Yeosu, Pohang, Jeju	*Paralichthys Olivaceus*, *Mugil cephalus*	1.7-28.8	ng/g wet-wt.	This study

*ND: Not detected

usually inhabits marine sediments where OTCs are detected in relatively higher concentration, had relatively higher concentration of OTCs. On the other hand, the concentration of OTCs detected in the muscle of the two fish species was similar to that detected in other fish species, and it is determined that the exposure of OTCs from the intake of the muscle is small.

Materials and Methods

Sampling

An analysis was conducted on olive flounder and flathead mullet which are popular commercial fish inhabiting coastal regions of Korea. The samples were collected between November 2001 and April 2002 from six coastal regions in Korea where large harbors are located: 5 samples from live-fish market in each region. After dissecting them, the muscles and livers from these samples were used for analysis (Figure 1). Extracted tissues were gathered together for each region and were homogenized by using homogenizers and analyzed. The length of the analyzed olive flounder was 28.5-51.0 cm and that of the flathead mullet was 32.5-46.0 cm. All samples were kept at lower than $-20^{\circ}C$ before they were analyzed.

Analysis Methods

OTCs were analyzed according to Analytical Methods of Endocrine Disrupting Chemicals[29] and Maritime

Environment Pollutant' Testing Method[30]. All the organic solvents used for the extraction were of a pesticide grade and the reagents were of a special grade. And, glassware were purged over a strong heat of 420 °C for more than 3 hours in order to prevent cross-contamination.

About 2 g of bio-samples was added with tripentyltin chloride (AccuStandard, USA), which is a surrogate chemical standard, and with a 0.1% troporone/benzene and 1 N HBr/ethanol. After the samples were ultrasonically extracted and centrifuged, the suspension was taken into an erlenmeyer flask, and this operation was repeated once again. The suspension was washed by adding 2 M NaBr/water, and benzene layer was separated from the suspension. The sample was then concentrated by using a rotary vacuum evaporator (Eyela, Japan) and was added with methanol/hexane (1 : 1), so as to remove fat and gain the separated methanol layer. This operation was repeated once again to obtain methanol, which was replaced with benzene and then derivatized with 27% propyl magnesium bromide/THF (TCI, Japan). In about 30 minutes the sample was added with a saturated ammonium chloride solution to stop the reaction. After the sample was centrifuged, the benzene layer was separated from it and was concentrated by using a rotary vacuum evaporator, and afterwards it was put through a column filled with florisil and silicagel for refining. The refined solution was added with tetrapentyltin chloride (AccuStandard, USA), which is an internal standard solution for confirmation, and finally concentrated into 200 μL with nitrogen gas in order to obtain a sample solution for measurement. Quantitative and qualitative analyses of the sample required repeated use of equipment such as gas chromatograph (GC) and mass spectrometer (MS) (Shimazu, GC 17A, QP-5050A). Also, J&W DB-5 column (30 m × 0.25 mm × 0.25 μm) (Agilent, USA) was used, and the sample was injected in the splitless mode. The detection was made using SIM mode, which is a method to select and detect ions of specific mass only. The analytical conditions of the instruments for OTCs are listed in Table 1.

In order to know the detection limit of OTCs, by adding an organotin standard whose detection limit was five times as much as the instrument detection limit (ILD) to bio-sample, the five samples that had been passed though the same pre-treatment process as the bio-sample were analyzed. Standard deviation was derived from the earned instrument measurement values and multiplied by 1.943 at the level of 95% to calculate the detection limit whose value was less than 0.1 ng/g. The recovery rate of OTCs was measured by adding about 2 g of bio-sample to 0.5 μg of each standard substance using the same method used in the analysis of the sample. The recovery rates were 80.0-97.5 % for MBT, 85.1-100% for DBT, 84.6-100% for TBT, 74.4-94.9% for MPT (Monophenyltin), 81.3-97.3% for DPT (Diphenyltin) and 86.4-118.6% for TPT (Triphenyltin), which were considered satisfactory; meanwhile, the relative standard deviation as well showed a reliable reproducibility at 5.4-10.6%.

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