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Accumulation of Butyltin Compounds in Shellfish and Fish from Korean Coastal Areas

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Butyltins (BTs) were measured in 2 species of shellfish and 16 species of fish collected along the Korean coast. Tributyltin (TBT) was detected in the mussels and oysters of sentinel organisms (20 to 940 ng Sn/g dry wt), indicating widespread contamination of TBT in the Korean coast. The elevated concentrations of TBT in the shellfish found in the sites near harbors or shipyards suggested that antifouling paints are probable major sources of butyltins in these areas. The TBT compound was detected in 12 out of 16 fish samples. The concentrations of TBT in fish muscles were between 7 and 151 ng Sn/g dry wt, while the level in whole body of anchovy was very high (793 ng Sn/g dry wt). Exposure doses to Korean people via consumption of these marine products were evaluated. The results suggested that the environmental levels of TBT were below the level of concern.

Key words: Butyltins, Shellfish, Fish, TBT, Exposure doses

INTRODUCTION

Since the mid-1960s, tributyltin (TBT) has been used in antifouling paints as an effective biocidal agent to keep ship hulls free from algae, barnacles and other fouling organisms. However, marine pollution by TBT has become a matter of great concern because of its lipophilic property and toxicity, particularly with regards to mollusks, even at low environmental concentrations (Evans, 1999). The first evidence of the harmful effects of TBT was found in oysters in France in 1982 (Alzieu *et al.*, 1991). Tributyltin also induces imposex on gastropod mollusks. Imposex is the superimposition of male sexual organs (penis and vas deferens) on female gastropods, which may bring about reproductive failure and consequential population decline in some species (Bryan *et al.*, 1986). Tributyltin produces deleterious effects even in extremely low concentrations. For example, concentrations of TBT less than 0.5 ng/l have been shown to cause widespread cases of imposex in the dog whelk (Bryan *et al.*, 1986; Gibbs *et al.*, 1991). Recognition of the environmental risks associated with TBT use has resulted in restrictions in many countries. The Marine Environmental Protection Committee (MEPC) of the International Maritime Organization (IMO) requires that TBT-based antifoulants be totally banned from the year 2003 onwards (Christen, 1999; Evans *et al.*, 2000). Butyltins have been detected in tissues of marine animals worldwide (Kannan and Falandysz, 1997; Harino *et al.*, 1998; Hung *et al.*, 1998; Davis *et al.*, 1999; Elgethun *et al.*, 2000). In Korea, the use of TBT-based paints on fishery facilities and small vessels was banned from 1999. Recently, high concentrations of TBT have been shown to exist especially in sediments of harbors and shipyards in Korea (Hwang *et al.*, 1999; Shim *et al.*, 1999). Although the ban has been successful in reducing TBT levels in seawater (Alzieu *et al.*, 1986; Valkirs *et al.*, 1991), there is a need for continuous monitoring to understand TBT accumulation patterns in different fish and shellfish species. Bioaccumulation of TBT has been studied with the use of biomonitors for surveillance of aquatic environments, and seafood have been analyzed to prevent excessive exposure of TBT to humans (Wade *et al.*, 1988; Kannan *et al.*, 1995; Kanatireklap *et al.*, 1997; Kannan and Falandysz, 1997; Tanabe *et al.*, 2000). The purpose of this study is to determine the contamination levels of TBT in edible shellfish and fish immediately after the ban on the use of TBT antifouling paints in Korea and to evaluate the risk of TBT exposure to humans via con-

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*Corresponding author: hgchoi@nfrdi.re.kr sumption of these contaminated marine foodstuffs.

MATERIALS AND METHODS

Sampling and Analysis

Shellfish specimens were collected at 20 stations throughout the coastal areas of Korea during May to July 2001, and 16 fish specimens were purchased at the local markets along the East, South and West Sea of Korea during July to August 2001. The biometrics of the samples are given in Tables 1 and 2. Due to the geographical distribution of shellfish, mussels were collected in the South and East Sea and oysters were collected in the West Sea. The samples were immediately transported to the laboratory in a cooler box with ice or dry ice. All shellfish, anchovy and white-saddled reef-fish samples were composites of 30 to 60 whole organisms. The number of fish samples analyzed, except anchovy and white-saddled reef-fish, ranged from 1 to 10. The shells of the shellfish were removed and the whole soft tissues were pooled and homogenized. The muscles of 14 fish specimens and the whole bodies of 2 fish specimens (anchovy and white-saddled reef-fish) were homogenized to prepare composite samples. Samples were stored at −20°C and later freeze-dried. The analytical procedure was performed after combining and modifying the procedures of Wade *et al.* (1988) and Harino *et al.* (1992). The freeze-dried samples (2 g to 5 g) were extracted twice by shaking for 3 hrs with 20 ml of 0.1% tropolone-methylene chloride with the addition of 10 ml of 50% HCl in 50 ml centrifuge tube. Tripentyltin chloride was spiked before extraction as surrogate standard (1.0 mg). The total organic extract was concentrated to nearly dryness using a rotary evaporator. Each extract was made 5 ml with *n*-hexane and then propylated with 2 ml of Grignard reagent (*n*-propylmagnesium bromide). The remaining Grignard regent was decomposed with 10 ml of 1N H_2SO_4. The organic fraction was decanted and the aqueous fraction extracted with 10 ml of *n*-

Table 1. The range of length, height and weight of the shellfishes collected from Korean coastal areas

Area	Length (mm)	Height (mm)	Weight (g)	Area description
Mussel *(Mytilus edulis)*				
East Sea				
Sokcho coast	85–136 (116)[a]	32–56 (45)	16–50 (33)	near breakwater
Jumunjin coast	76–104 (90)	24–38 (32)	4–21 (10)	entrance of harbour
Jukbyeon coast	60–99 (75)	22–39 (30)	5–14 (9)	near Buku-ri
Hupo coast	73–108 (89)	29–39 (34)	9–18 (14)	near harbor
Gulyongpo coast	64–94 (79)	22–39 (30)	5–26 (11)	entrance of harbour
Ulsan Bay	28–43 (38)	10–17 (14)	1–3 (2)	near Ulsan harbour
South Sea				
East of Geojedo	7–69 (43)	9–25 (16)	1–10 (4)	near Okpo harbour
Busan Bay	42–93 (57)	13–28 (19)	2–19 (8)	near North harbour
Haengam Bay	31–46 (41)	10–19 (15)	1–5 (3)	entrance of harbour
Masan Bay	40–63 (50)	13–28 (19)	3–14 (6)	near Myodo
Gohyeon Bay	50–70 (61)	18–28 (23)	4–11 (7)	near harbour
Gwangyang Bay	63–84 (74)	31–50 (38)	5–19 (11)	near industrial complex
Gamak Bay	62–83 (73)	32–43 (38)	7–15 (10)	near aquaculture area
Yeongsan esturary	45–62 (51)	17–33 (21)	4–10 (6)	near harbour
Oyster *(Crassostrea gigas)*				
West Sea				
Gochang coast	38–63 (46)	16–28 (22)	1.0–4.5 (2.8)	near Yeongkwang atomic power plant
Jeonjupo coast	35–79 (51)	17–35 (23)	1.1–10.9 (3.5)	near Seonyudo
Gunsan coast	33–64 (47)	16–34 (24)	0.8–5.4 (3.0)	near Osikdo
Cheonsu Bay	28–53 (43)	22–35 (28)	1.2–2.0 (1.6)	near Cheonbukmyeon, Bolyeong
Asan Bay	25–43 (34)	16–29 (21)	1.4–2.4 (1.6)	near industrial complex
Incheon coast	34–67 (48)	18–32 (27)	1.2–2.2 (1.8)	near harbour

[a]Numbers in parenthesis represent mean values.

Table 2. Main characteristics of fishes purchased at local markets along Korean coastal areas

Local Market	Species	n[a]	Length (cm)	Height (cm)
East Sea				
Gangleung	Roundnose flounder (*Eopsetta grigorjewi*)	10	20	8
Pohang	Saury (*Cololabis saira*)	8	28	3
Pohang	Herring (*Clupea pallasi*)	6	27	6
South Sea				
Yeosu	Sharp toothed eel (*Muraenesox cinereus*)	10	33	4
Yeosu	Red tongue sole (*Cynolossus joyneri*)	10	14	9
Yeosu	Sea Bass (*lateolabrax japinicus*)	1	38	11
Yeosu	Silver Fish (*Pampus argenteus*)	7	19	10
Mokpo	Flatfish (*Paralichthys olivaceus*)	8	43	16
Mokpo	Jacopever (*Sebastes schlegeli*)	3	30	10
Tongyeong	Anchovy (*Engraulis japonica*)	60	4	1
Tongyeong	Common conger (*Conger myriaster*)	10	32	3
Jeju	Whitesaddled reeffish (*Chromis notata*)	50	5	3
Busan	Hairtail (*Trichiurus lepturus* Linnaeus)	5	31	8
Busan	Mackerel (*Scomber japonicus*)	5	42	6
West Sea				
Incheon	Greening (*Hexagrammos otakii*)	10	25	5
Gunsan	Pomfret (*Pampus echinogaster*)	8	20	10

[a]Number of individuals

hexane. The combined organic phase was concentrated by a rotary evaporator and then purified by being passed through a 10 g Florisil packed wet column. In the fish samples with high contents of lipid, the extract was passed through a 20 g Florisil packed dry column and then purified by being passed through a 10 g Florisil packed wet column. The extract was eluted with 15 ml of n-hexane. The organic part of eluate was concentrated to 1 ml under N_2. Finally, the internal standard (tetrabutyltin) was added in the concentrated eluates to evaluate the recovery. The butyltins level was determined on a Hewlett-Packard 6890 gas chromatograph (GC) equipped with a flame photometric detector with a 610 nm cut-off interference filter. A capillary column HP 5 was used (5% phenyl methyl siloxane, 30 m×0.25 mm internal diameter ×0.25 μm film thickness). The temperature program was set as follows: initial at 80°C, 80−160°C at 15°C/min, 160−200°C at 5°C/min, and isothermal 220°C for 3 min. The injector and detector temperatures were set at 220°C and 200°C, respectively. Nitrogen was used as a carrier and make-up gas at flow rates of 1.4 ml/min and 18.6 ml/min, respectively. Hydrogen and air were passed at 150 ml/min and 100 ml/min, respectively. Values were given in ng/g dry wt as Sn. Quality assurance and quality control procedure included internal standards, procedural blanks, the analysis of a reference material (NIES No 11; Sea bass, Japan) and spiked samples. The recovery in the reference material was more than 90% for TBT. The recovery of the spiked sample (red-tongue sole) was 101±2% for TBT, 99±23% for dibutyltin (DBT) and 105±27% for monobutyltin (MBT). Detection limits were around 3 ng Sn/g dry wt for TBT, 5 ng Sn/g dry wet for DBT and 4 ng Sn/g dry wt for MBT. Lipid was measured using automatic extraction unit (Gerhardt and Variostat, Germany) with n-hexane.

Exposure and Risk Assessment

The daily TBT exposure dose (ng/kg/day) from seafood consumption was estimated using a standard exposure model (USEPA, 1989; Sydney water Corporation, 1995; Robinson *et al.*, 1999) as follows:

$$\text{Exposure}=\frac{\text{CS}\times\text{IR}\times\text{EF}\times\text{ED}}{\text{BW}\times\text{AT}}$$

where CS is the concentration of TBT (ng/g) in the seafood, IR is the seafood ingestion rate (g/day), EF is the exposure frequency (365 days/year), ED is the exposure duration (30 years), AT is the averaging time (exposure duration ×365 days/year) and BW is the body weight (60 kg).

The potential risks were quantified as a hazard quotient (HQ) by comparing the exposure dose from seafood to TBT Tolerable Daily Intake (TDI) or TBT Acceptable Daily Intake (ADI) as follows:

$$\text{Exposure dose}=\frac{\text{Exposure dose (ng/kg/day)}}{\text{TDI or ADI (ng/kg/day)}}$$

RESULTS AND DISCUSSIONS

BTs in Shellfish

The levels of BTs along with the lipid content in the mussels and oysters are presented in Table 3. The lipid content varied from 4.6 to 10.2% (dry weight basis) in mussels and from 6.3 to 12.9% (dry weight basis) in oysters. The ranges of TBT concentrations were 40 to 186 ng Sn/g dry wt in oysters and 20 to 940 ng Sn/g dry wt in mussels. The concentrations of TBT in the mussels varied widely depending on the sampling location, while those of oysters showed a similar distribution throughout the sampling sites. High concentrations of TBT in mussels were found in Ulsan, Hupo and Busan coastal areas surrounded by industrialized cities and harbors. The TBT levels in the mussels from Ulsan Bay, in particular, exceeded the threshold value of the effects on scope for growth of mussels (816 ng Sn/g dry wt) (Page and Widdows, 1991). These levels were comparable to those in mussels from the Pacific coast of the United States (Short and Sharp, 1989) and central-west Greenland (Jacobsen and Asmund, 2000). However, these levels were considerably lower than those in *Gafarium tumidum* tissue (4,286 ng Sn/g dry wt) taken from Lami Dump (Davis *et al.*, 1999). It is well known that the antifouling paints leached from boats and vessels are major contributors of butyltins to the marine environment (Schatzberg, 1987; Alzieu, 1998; Tanabe *et al*,

Table 3. Concentrations of butyltin compounds (ng Sn/g dry wt) in the shellfish from Korean coastal areas

Areas	Lipid (%)	MBT	DBT	TBT	Σ BTs[a]	TBT/ ΣBTs (%)
Mussel *(Mytilus edulis)*						
East Sea						
Sokcho coast	4.6	72	93	124	289	43
Jumunjin coast	4.7	19	20	30	69	44
Jukbyeon coast	4.6	19	20	20	58	34
Hupo coast	4.8	135	230	497	862	58
Gulyongpo coast	8.6	25	32	28	85	33
Ulsan Bay	10.1	785	962	940	2687	35
South Sea						
East of Geojedo	7.0	392	181	117	691	17
Busan Bay	6.0	215	290	451	956	47
Haengam Bay	10.2	39	56	54	149	36
Masan Bay	6.8	51	45	23	119	19
Gohyeon Bay	5.1	138	175	113	426	27
Gwangyang Bay	7.0	43	36	31	110	28
Gamak Bay	9.1	64	34	33	131	25
Yeongsan Estuary	7.2	167	80	104	351	30
Oyster *(Crassostrea gigas)*						
West Sea						
Gochang coast	11.7	27	32	119	178	67
Jeonjupo coast	6.3	nd[b]	15	40	54	73
Gunsan coast	12.9	25	24	162	211	77
Cheonsu Bay	9.3	36	31	186	254	73
Asan Bay	10.3	12	17	122	151	81
Incheon coast	8.0	38	22	125	185	68

[a] ΣBTs=MBT+DBT+TBT

[b] nd=not detected

2000). Therefore, TBT contamination is concentrated especially along the harbors, shipyards and the places where boating activities are high. All the shellfish contained TBT and its metabolites, DBT and MBT. The contribution of TBT to total BTs ranged from 17 to 58% in the mussels and from 67 to 81% in the oysters. Harino *et al.* (1998) reported that the ratio of TBT to total BTs in the mussel of Otsuchi Bay ranged from 52% to 68%, showing a decrease with an increase in distance from the shipyard. The ratios of oysters were comparable to those in the Coos Bay estuary of Oregon, USA (Elgethun *et al.*, 2000). The concentrations of butyltins found in bivalves reflected the time-integrated water column concentration and the bivalves rate of uptake, metabolism, and depuration of individual butyltin compounds (Wade *et al.*, 1988). The complexity of the interaction of these processes was reflected in the various proportions of TBT, DBT, and MBT found in these bivalves.

BTs in Fish

Tributyltin was detected in 12 of the 16 fish specimens (Table 4). The TBT concentrations in the fish samples ranged from 7 to 793 ng Sn/g dry wt, depending on sampling location and species. The highest TBT levels were detected in anchovy, whereas the TBT levels in red-tongue sole, white-saddled reef-fish, hairtail and mackerel were below the detection limits. The considerably elevated level of TBT in anchovy may be an outcome of difference in sampling part of the fish for analysis. Anchovy was analyzed as whole body and the other samples analyzed as muscles. Guruge and Tanabe (2001) also reported that the BT concentrations in the liver of rabbit fish were an order of magnitude higher than those in muscle and eggs, suggesting the liver may be the target organ in fish. The concentration of TBT in anchovy was comparable to those in deep-sea organisms and relatively lower than those in shallow-water fish from Suruga Bay of Japan (Takahashi *et al.*, 1998). The concentrations of DBT and MBT varied depending on the species. Relatively high contributions of MBT were found in flatfish, roundnose flounder, jacopever, common conger and greening, while high TBT levels were found in pomfret, silver fish, sharp-toothed eel and anchovy. In particular, sea bass, saury and herring flesh showed no metabolites. The presence of less-toxic DBT and MBT indicates that degradation processes of TBT occur in fish. Takahashi *et al* (1998) reported that the environmental feature and/or the metabolic capacity may be likely causes for the dif-

Table 4. Butyltin concentrations (ng Sn/g dry wt.) in the fish from Korean costal areas

Local Market	Species	moisture (%)	lipid (%)	MBT	DBT	TBT	ΣBTs[a]	TBT/ΣBTs(%)
East Sea								
Gangleung	Roundnose flounder	83	7	43	7	151	202	75
Pohang	Saury	79	15	nd[b]	nd	26	26	100
Pohang	Herring	77	47	nd	nd	122	122	100
South Sea								
Yeosu	Sharp toothed eel	81	14	nd	13	35	48	73
Yeosu	Red tongue sole	76	2	nd	nd	nd	nd	-
Yeosu	Sea Bass	71	15	nd	nd	36	36	100
Yeosu	Silver Fish	81	18	nd	20	138	158	87
Mokpo	Flatfish	76	1	nd	8	19	26	70
Mokpo	Jacopever	80	5	243	nd	30	273	11
Tongyeong	Anchovy	77	12	504	130	793	1427	56
Tongyeong	Common conger	81	32	57	nd	34	91	38
Jeju	Whitesaddled reeffish	75	13	116	nd	nd	116	-
Busan	Hairtail	73	19	51	27	nd	191	-
Busan	Mackerel	68	48	nd	nd	nd	nd	-
West Sea								
Incheon	Greening	80	6	61	nd	7	68	10
Gunsan	Pomfret	84	15	53	23	140	217	65

[a] ΣBTs=MBT+DBT+TBT

[b] nd=not detected

ference of the ratios of TBT to total BT between deep-sea organisms and shallow-water organism. On the other hand, Guruge and Tanabe (2001) suggest that the habitat of fish may be the main factor for TBT contamination in marine organisms because the maritime activities and ship-repairing in the harbor is the main source for BT contamination. It is therefore very difficult to compare the fish data from different areas because the butyltin burden in fish depends on the species, habitat, metabolism and etc. The lipid contents varied between 1.2% in flatfish and 48.3% in mackerel (dry weight basis). Although considerable variation was observed in organochlorine (OC) concentrations among species, those levels are basically correlated with their body lipids. In contrast to OC, the bioaccumulation of TBT is not readily related to its lipophilicity because of its ionic nature (Yamada and Takayanagi, 1992; Takahashi *et al.*, 1998). In this study, a significantly lower lipid-dependent accumulation of BTs in organisms was observed.

Seafood Risks

Oysters and mussels are common types of edible shellfish in Korean coastal waters and the fish species analyzed here are commercially vital food items in Korea. Considering the toxic effects of butyltins, the extent to which contaminated fish may contribute to the human food chain is of considerable concern. The human health risks associated with the consumption of TBT-contaminated fish and shellfish were evaluated using the results in this study. As indicated in Table 5, TBT exposure doses estimated assuming bodyweight for an adult of 60 kg were 6 to 822 ng/kg/day (mean 79 ng/kg/day) for fish and 10 to 474 ng/kg/day (mean 84 ng/kg/day) for shellfish. Those exposure doses were below the ADI based on the acceptable daily intake (ADI) of 1600 ng TBTO/kg/day suggested by the Ministry of Health and Welfare in Japan (Yamada, 1999). However, EPA revised the oral reference dose (RfD) for TBTO to 280 ng/kg/day in 1997 (Elgethun *et al.*, 2000). According to this guideline, the upper values of the shellfish and fish exceeded the RfD of EPA with hazard quotients (HQ) of 1.693 and 2.936, respectively. For non-carcinogenic effects, the risk is expressed as a HQ, the ratio between exposure and ADI. An HQ value of more than 1 means that people who are exposed to contaminated mussels have potential health risks. However, Robinson *et al.* (1999) reported that there is a question associated with the approach used maximum concentrations. It is unrealistic to assume that an individual could consume the maximum concentration each day of the year over an indefinite period and therefore estimations using either a mean concentration (European Commission, 1996) or an upper confidence limit on the mean (Sydney Water Corporation, 1995) is more reasonable. Based on this approach, the exposure dose calculated through mean TBT level was below the oral reference dose of EPA.

CONCLUSION

This exploratory survey suggests that TBT is a widespread contaminant in the marine organisms of Korean coastal areas. The concentrations of TBT in the mussels were high in coastal areas surrounded by industrialized cities and big harbors. The concentrations of TBT in muscles of fish from Korean coastal areas were comparable to those from other countries, except for whole-body anchovy, which showed higher TBT levels. The results indicate that the levels of TBT in the fish composed of different species have no correlation with lipid contents. The contributions of TBT to the total butyltin burden var-

Table 5. Risk assessment from marine products in Korean coastal areas

Species	Per caput supply[a] (g/day)	TBTO Conc. (ng/g wet wt)	Exposure dose (ng/kg/day)	ADI (ng/kg/day)	HQ[b]
Fish	53.89	7-915 (88)[c]	6-822 (79)	1600[d]	0.004-0.514 (0.049)
				280[e]	0.021-2.936 (0.281)
Shellfish	30.21	20-942 (167)	10-474 (84)	1600[d]	0.006-0.296 (0.053)
				280[e]	0.036-1.693 (0.300)

[a]Data was quoted from Food Balance Sheet of Korea Rural Economic Institute
[b]Hazard quotient=exposure dose/ADI
[c]( )=mean value
[d]Ministry of Health and Welfare in Japan
[e]the oral RfD of EPA

ied among fish species and ranged from 17 to 81% in shellfish and from under the detection limit to 100% in fish. The human health risks estimated from the results in this study indicate that exposure dose was below the ADI of Japan and the oral RfD of EPA.

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