Korean J. Environ. Biol. 22 (Special issue) : 63~70 (2004)

Tributyltin Compound in Sediments and Tissues of Oysters and Rock Shell in Gwangyang Bay, Korea

Won Joon Shim*, Un Hyuk Yim, Nam Sook Kim, Sang Hee Hong and Jae Ryoung Oh

Southern Coastal Environment Research Division, South Sea Institute, KORDI, 656-834, Korea

Abstract - Tributyltin (TBT) and its degradation products, dibutyltin (DBT) and monobutyltin (MBT) were quantitatively determined in surface sediments and two molluscan species, Pacific oyster (*Crassostrea gigas*) and rock shell (*Thais clavigera*), from Gwangyang Bay, Korea. Butyltin compounds were detectable in almost all sediment and biota samples. Tributyltin concentrations in surface sediment ranged $<2\sim33$ ng g^{-1}, which is at a lower end of TBT concentrations in industrialized bays in Korea. However, TBT levels in sediments were related to boating activities around the bay. In biota samples, TBT concentrations were in the range of $178\sim2{,}458$ ng g^{-1} for oyster and $47\sim236$ ng g^{-1} for rock shell. Relatively high TBT concentrations in biota were found near wharves for fisherboats and harbor areas. About $90\sim100\%$ of the female *T. clavigera* displayed imposex, and relative penis length index of the imposexed-female was in the range of $20.9\sim107.9\%$. Furthermore, TBT body residue had a significant positive relationship with degree of imposex in *T. clavigera*. Overall, TBT concentrations in Gwanyang Bay were much lower than other major bays in Korea.

Key words : Tributyltin, Sediment, Oyster, Imposex

INTRODUCTION

Organotin compounds are among the most-widely used organometallic chemicals. Production of organotins has increased for various purposes since their first biocidal application in the early 1920s (Thompson *et al.* 1985). One of the major applications lies in heat stabilization for synthetic polymers and another lies in their use as agricultural biocide (WHO 1990). Tributyltin (TBT) used as biocidal additive for antifouling paints has received much attention since its adverse effects on oyster farms near marina were revealed in the early 1980s (Alzieu 1986). A series of studies have showed that TBT is highly toxic toward various non-target marine organisms (for review, see Fent 1996). Due to the induction of imposex (imposition of male sexual organs on females) in gastropod species, TBT was classified as endocrine disruptor (Oehlmann *et al.* 1996; Horiguchi *et al.* 1997). As imposex is mainly caused by TBT, it is widely used as a biomarker for detecting effects of TBT on marine organisms. Many previous studies have shown positive dose-response relationships between TBT concentrations and degree of imposex in neogastropod species (Gibbs *et al.* 1987; Stroben *et al.* 1992; Shim *et al.* 2000).

Starting in the 1980s, many industrialized countries have regulated the use of TBT in antifouling paints. The application of trialkyltin-based antifouling paints on ships except for ocean-going vessels, nets and immersed structures has been banned in Korea, since 2000. However, there are no regulations for releasing of wastewater containing TBT from shipyards in Korea yet. Furthermore, no regulations are implemented in most

* Corresponding author: Won Joon Shim, Tel. 055-639-8671, Fax. 055-639-8689, E-mail. wjshim@kordi.re.kr

of the Asian countries. Even though high concentrations of TBT were found in Aisan coastal environments (Kannan *et al.* 1995; Kan-atireklap *et al.* 1997; Shim *et al.* 1998; Sudaryanto *et al.* 2002; Shim *et al.* 2002), its effects upon marine life have not yet been well documented.

In the present study, TBT and its degradation products in sediment and biota (oyster and rock shell) samples collected from Gwangyang Bay were quantitatively determined. The spatial distribution of butyltin compounds in surface sediments and oysters was measured to investigate the extent of contamination and major sources of TBT in throughout the bay. Bioaccumulation of TBT compound and degree of imposex in gastropod were evaluated to understand its effects on marine organism in the bay.

MATERIALS AND METHODS

1. Study area and sample collection

Gwangyang Bay is located at the southern coast of Korea (Fig. 1). The inner bay is classified as industrialized one, including a large steel mill plant in the middle of northern part of the bay, petroleum refinery complex in southern part of the bay. There are many harbor facilities including quays and piers for ocean going vessels and several small wharves for fishing boats around Gwangyang Bay. In addition, almost two-third of coastal lines of the inner bay have been modified by human activities.

Sediment samples were collected from 17 stations using a van Veen grab in June, 2001 (Fig. 1). Approximately the top 2 cm of sediments were removed using a stainless steel spoon and stored in pre-combusted amber bottles. Oyster (*Crassostrea gigas*) samples were collected at 9 sites (Sites B1-B4, B6, B8, B12, B15, and B16) around the bay from September to October, 2001 (Fig. 1). Rock shell (*Thais clavigera*) were collected at 11 sites (Sites B5~B15) from the estern part of the inner bay and the outer bay from November 2001 to January 2002 (Fig. 1). Feral oyster and rock shell samples were collected during low tide depending on places where the target species inhabitate. Rock shell is hard to find in the western part of the inner bay. The sediment and

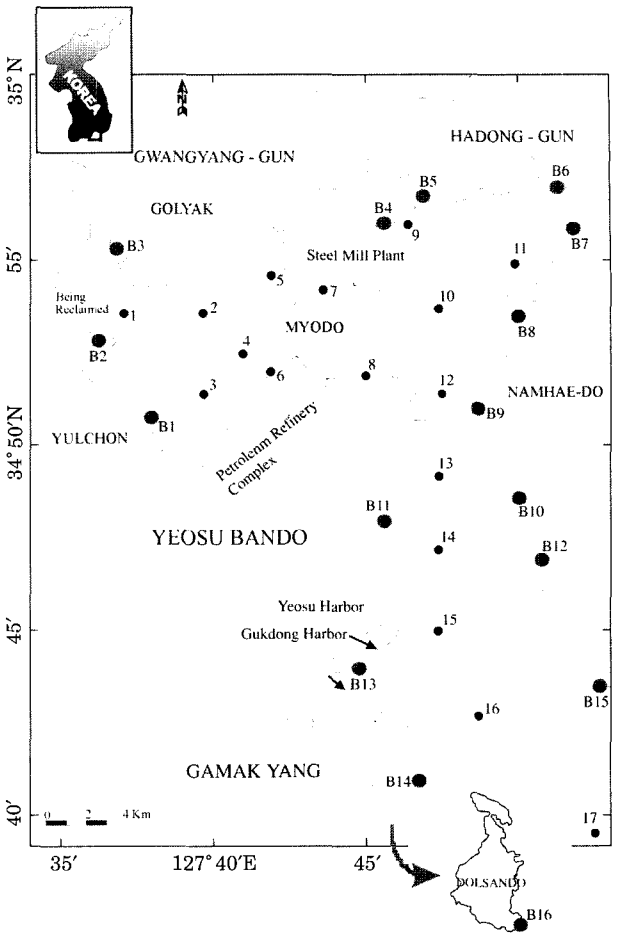

Fig. 1. Location map of surface sediment (small circle) and biota (large circle) sampling sites in Gwangyang Bay.

biota samples were frozen immediately with dry ice and taken to the laboratory for analysis.

2. Analytical procedure for organotins

Organotin analysis was performed by gas chromatography equipped with a flame photometric detector (GC-FPD) after propylation in aqueous phase with propylmagnesium bromide, using a previously described method (Shim *et al.* 2002; Shim *et al.* 2003). Twenty individuals of oyster or rock shell from each site were pooled and homogenized in a glass amber bottle. Tripentyltin chloride was added to the samples as a surrogate standard. The samples were digested with 10 ml of 6 N HCl for 30 min, and then extracted with 20 ml dichloromethane with tropolone (0.1%) by shaking for 3 h. The

Table 1. Comparison of certified and measured values (n = 6) of butyltins in certified reference material (CRM477) in mussel tissue

Concentration (μg g^{-1} dry weight as cation)			
Organotin speices	Monobutyltin	Dibutyltin	Tributyltin
Mean±SD	1.59±0.04	1.31±0.03	1.74±0.04
Certified value	1.50±0.28	1.54±0.12	2.20±0.19

Table 2. Comparison of certified and measured values (n = 6) of butyltins in certified reference material (PACS-1) in sediments

Concentration (μg g^{-1} dry weight as Sn)			
Analyte	Monobutyltin	Dibutyltin	Tributyltin
Mean±SD	0.58±0.07	1.09±0.03	1.23±0.03
Certified value	0.28±0.17	1.16±0.18	1.27±0.22

derivatized extracts were recovered by centrifugation and cleaned up on 1 g of activated Florisil (Supelco, Bellefonte, PA, USA). The cleaned extracts were concentrated again and spiked with tetrabutyltin as an internal standard, and then analyzed by GC-FPD.

For sediment, freeze-dried sediments were extracted with the same method for the tissue samples. The sediment extracts were cleaned-up using 2 g of activated Florisil.

The gas chromatograph (GC) operating condition was identical to that in Shim *et al.* (1998) except for the temperature program. The GC temperature was programmed from 50°C (2 min holding time) and heated to 150°C (10 min holding time) at a rate of 10°C min^{-1}, and then heated again to a final temperature of 240°C (5 min holding time) at 30°C min^{-1}.

The whole analytical procedure is validated by analyzing reference materials for tissues from BCR, EU (CRM477 mussel tissue) and sediment from NRC, Canada (PACS-1), and their results are given in Tables 1 and 2. The analytical results of the reference materials fell within the range of the certified or close values except for MBT in sediment, which was two times higher than the certified values. However, recent studies have reported similarly 1.5~2 times higher MBT values of PACS-1 and another reference material, PACS-2, than certified values (Moens *et al.* 1997; Roderiguez *et al.* 1999).

Recoveries of butyltin compounds from spiked (0.1 μg Sn g^{-1}, n = 7) oyster tissue and sediment samples were in the range of 85~110% and 84~117%, respectively. Relative standard deviation of the butyltin spiked samples were less than 4.7% for oysters and less than 7.7% for sediments. The method detection limits of the whole analytical procedure for tissue and sediments samples ranged from 2 to 5 ng g^{-1}. Concentrations of organotin compounds in sediment and tissue samples are expressed as ng g^{-1} of Sn on a dry weight basis to allow direct comparisons of the organotin compounds.

3. Measurement of imposex

About 20 to 30 rock shell from each site was used to measure frequency and degree of imposex. Shell length was measured with Vernier calipers. The shells were cracked open with a titanium tool and the soft tissue were removed. Sex of each gastropod was determined by appearance of a prominent female sperm-ingesting gland (Horiguchi *et al.* 1994). The penis of both female and male were removed, and penis length was measured to the nearest 0.1 mm using Vernier calipers. The frequency of imposex females was expressed as a percentage. Whereas the degree of imposex was expressed by index of relative penis length (RPL): [(mean length of female penis)/(mean length of male penis) × 100] (Bryan *et al.* 1986).

RESULTS AND DISCUSSION

1. Occurrence and spatial distribution of butyltins in sediments

Tributyltin (TBT) and its degradation products, DBT and MBT, were detected from 14 out of 17 sites surveyed (Fig. 2). Phenyltins, however, were below detection limits at all sites. The overall ranges of butyltins in sediments were from <2 to 33, <2 to 12, and <3 to 21 ng g^{-1} for TBT, DBT, and MBT, respectively. Concentrations of TBT had a significant correlation (r^2 = 0.67; $p < 0.05$) with DBT in sediments, while MBT showed no significant ($p > 0.05$) correlations with the other butyltins. The mean concentration of TBT, DBT, and MBT determined were 13, 6, and 11 ng g^{-1}, respectively. The concentration of TBT accounted for the largest portion (43%) of

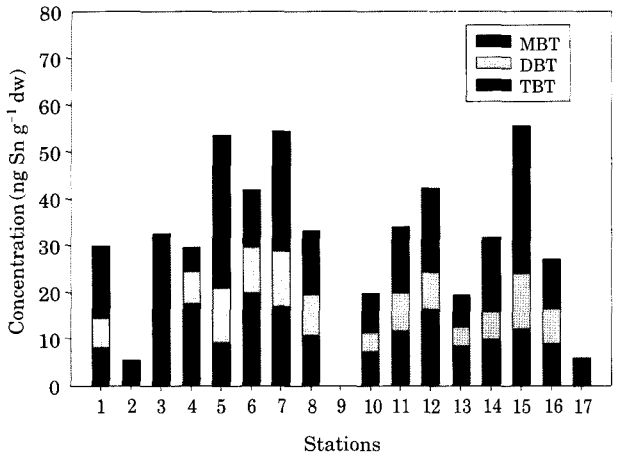

Fig. 2. Concentrations of three butyltin compounds in surface sediments collected from Gwangyang Bay.

Table 3. Comparison of tributyltin (TBT) concentrations in surface sediments collected from coastal areas of Korea

Location (number of data)	Concentration (ng Sn g^{-1} dw)		Reference
	Min~Max	Mean±SD	
Pohang Bay (n = 6)	15~2,730	613±1,057	MOMAF (1998)
Ulsan Bay (n = 10)	26~13,300	1,819±4,055	"
Busan area (n = 11)	16~2,360	355±683	"
Okpo Bay (n = 5)	495~11,700	5,379±4,921	"
Mokpo Bay (n = 10)	33~342	55±96	"
Gamak Bay (n = 10)	4~19	8±5	Shim (2000)
Gunsan area (n = 10)	ND[a]~21	7±7	"
Chunsu Bay (n = 9)	ND~4	0.4±1.3	"
Incheon North Harbor (n = 6)	16~375	86±142	"
Chinhae Bay (n = 60)	4~382	60±87	Shim *et al.* (1999)
Gwangyang Bay (n = 17)	ND~33	10±9	This study

[a]ND: not detected

total butyltin composition, and it was followed by MBT (38%) and DBT (18%). Overall TBT concentrations in sediments of Gwangyang Bay are much lower than those reported in the other harbor areas of Korea (MOMAF 1998; Shim *et al.* 1999; Shim 2000) (Table 3). Shim (2000) reported the similar range of TBT concentrations in Gamak Bay (4~19 ng g^{-1}) and Gunsan Harbor (not detected~21 ng g^{-1}) to those in Gwangyang Bay in this study (Table 3).

TBT levels were relatively high at Sts. 5 (33 ng g^{-1}) and 7 (26 ng g^{-1}) in front of the steel mill plant and at St. 15 (32 ng g^{-1}) near the Yeosu Harbor. Butyltin compounds were below detection limits at St. 9, mouth of Seomjin River. Even though none of distinct hot spot of TBT contamination was found in Gwangyang Bay, overall spatial distribution of total butyltin in the bay showed a source related tendency. Relatively high total butyltin concentrations were found at Sts. 3 through 8 (30~54 ng g^{-1}) in the inner part of the bay where quays and wharves for large vessels are located. At St. 11, near which several wharves for fisherboats are located, demonstrated a slightly higher total butyltin concentration (42 ng g^{-1}) compared to those at Sts. 9, 10, and 12 (not detected~34 ng g^{-1}) which are on the lines between Seomjin River mouth and Gwangyang waterway. Below detection limit of TBT in Seomjin River mouth indicates that TBT contaminations are originated not from land but from marine environment. A negative gradient of total butyltin concentrations from St. 15 (57 ng g^{-1}), the nearest station to Yeosu Harbor and Gukdong Harbor, to Sts. 14 and 13 (31 and 19 ng g^{-1}) and Sts. 16 and 17 (27 and 6 ng g^{-1}) indicates that butyltin are introduced to Gwangyang waterway from those harbors. These distribution pattern of total butyltin concentrations in Gwangyang Bay implies that butyltin contamination is closely related to shipping activities. The primary source of TBT in Gwangyang Bay is thought to be antifouling agent leached from ship hulls as in other coastal areas (e.g., Dowson *et al.* 1992; Tolosa *et al.* 1992; Yonezawa *et al.* 1993).

When arrival number of ships at major harbors of Korea is considered, TBT concentrations in Gwangyang Bay are much lower than Busan Harbor (Table 3). According to the stastics reported by MOMAF (1997), the number of ships arrived at Busan Harbor (33,557) is two times higher than that of Gwangyang Bay (16,757) Harbors. In addition, tonnage of ships at Busan (205,216,729 MTs) is approximately two times higher than that of Gwangyang Bay (85,511,241 MTs) in 1997. However, considering the difference of TBT concentrations between Busan Harbor (355±683 ng g^{-1}) (MOMAF, 1998) and Gwangyang Bay in this study (13±9 ng g^{-1}), total number or tonnage of ships arrived at the bays could not fully account for the TBT levels found in Gwangyang Bay. This discrepancy is probably caused

by different number of small ships in both bays. Number of ships less than 100 MTs in Busan Harbor (1,628) is about an order of magnitude higher than that in Gwangyang Bay (117) in 1997. Leaching of TBT antifouling agent from ship hulls is enhanced when ships are in motion, and is influenced by paint types and surface to volume ratio of ships. Small ships, including many fisherboats move much more frequently in coastal areas than large ships do and have much higher surface to volume ratios. Furthermore, it is known that conventional type of TBT based paints which have higher leaching rate than self-polishing copolymer (SPC) type are mainly applied to small ships due to their two or three times lower price than SPC type paints.

The TBT concentrations in this study can be compared to the results of TBT survey in Gwangyang Bay, 1995 (Shim 2000). Although the sampling stations in the 1995 survy are confined to the inner part of the bay, approximately to St. 12 in this study, sediment TBT concentrations in 1995 (2~214 ng g^{-1}) are higher than those in 2001. The mean TBT concentration in 1995 (mean±sd = 20±37 ng g^{-1}) is about two times higher than that in 2001 (13±9 ng g^{-1}). The decrease of TBT concentrations in surface sediment from 1995 to 2001 is partly explained by reduction of input sources. The use of TBT based paint on ships less than 400 tonnage has been banned since 2000 in Korea. Total ban of using TBT based paint on ships by International Maritime Organization (IMO) is expected to be effective in 2003. Most of shipping activities in Gwangyang Bay are done by large vessels except for the eastern part of the inner bay, where fisherboat activity is dominant. However, because half-life of TBT in sediment was estimated up to several years (de Mora *et al.* 1989), further monitoring and fate study are required to evaluate efficiency of the TBT regulation in the coastal environment of Korea.

2. Butyltin residues in oyster

The concentrations of butyltins in oysters are shown in Figure 3. All the oyster samples analyzed from Gwangyang Bay contained detectable butyltins, but phenyltins were below detection limits in all samples. The overall ranges of butyltin concentrations in oyster were 178 to 2,458 ng g^{-1} (mean±sd, 561±721 ng g^{-1}) for

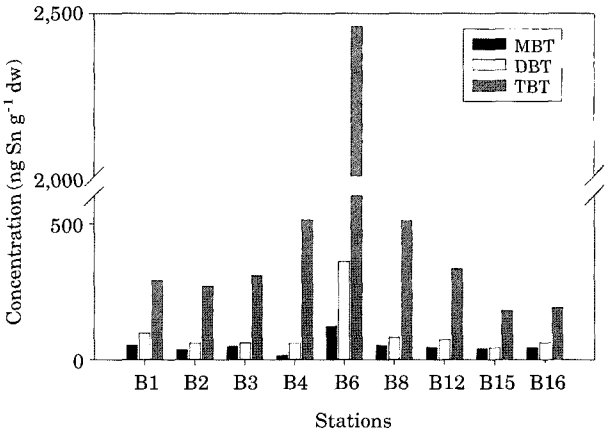

Fig. 3. Concentrations of three butyltin compounds in oyster (*Crassostrea gigas*) collected from Gwangyang Bay.

TBT, 42~359 ng g^{-1} (101±98 ng g^{-1}) for DBT, and 14 to 122 ng g^{-1} (51±29 ng g^{-1}) for MBT, respectively. The highest TBT concentration was observed at site B6 near wharves for fisherboats. The overall TBT levels in oyster were relatively high in the eastern part of the bay. DBT and MBT concentration in oyster showed similar distribution patterns to that of TBT. TBT and DBT concentrations were highly correlated ($r^2 = 0.96$; $P<0.05$) as were DBT and MBT ($r^2 = 0.77$; $P<0.05$), respectively. These correlations among butyltins demonstrate DBT and MBT are degradation products of TBT. TBT was major butyltin compound in oyster accounting for 74% of total butyltin concentrations.

The TBT concentrations in oyster from Gwangyang Bay in 2001 in this study can be compared to the results of TBT survey in Gwangyang Bay, 1995. Although the oyster sampling sites in the 1995 survy are not identical to those in this study, TBT concentrations in 2001 (561±721 ng g^{-1}) are comparable to those in 1995 (558±332 ng g^{-1}) (Shim 2000)

3. Occurrence and degree of imposex in rock shell

Occurrence and degree of imposex in *T. clavigera* collected from 11 sites on the coast of Gwangyang Bay are shown in Table 4. The frequency of imposexed-females was greater than 90% at at all sites surveyed. Relative penis length indices (RPLI) ranged from 20.9 to 107.9% (Table 4). The overall degree of imposex in the inner bay

Table 4. Summary of imposex and tributyltin (TBT) and tota. butyltin concentrations in gastropod, *Thais clavigera*

Site no.	No. of samples	FMPL[a] (mm)	MMPL[b] (mm)	% female of imposex	RPLI[c] (%)	TBT (ng Sn g^{-1})	BTs[d] (ng Sn g^{-1})
B5	25	9.95	12.26	100	81.2	111	710
B6	30	12.37	13.44	100	92.0	143	1,040
B7	30	13.25	12.28	100	107.9	139	935
B8	27	9.85	11.31	100	87.1	186	463
B9	30	6.28	10.64	100	59.0	122	519
B10	24	9.02	11.45	100	78.8	170	437
B11	25	8.22	10.42	100	78.9	80	395
B12	30	6.01	12.67	100	47.9	104	393
B13	20	12.60	12.30	100	102.3	236	2,717
B14	20	5.82	8.71	100	66.8	56	271
B15	27	2.16	10.34	90	20.9	47	144

[a]FMPL: female mean penis length
[b]MMPL: male mean penis length
[c]RPLI: relative penis length index
[d]BTs: total butyltin (sum of mono-, di- and tributyltin compounds)

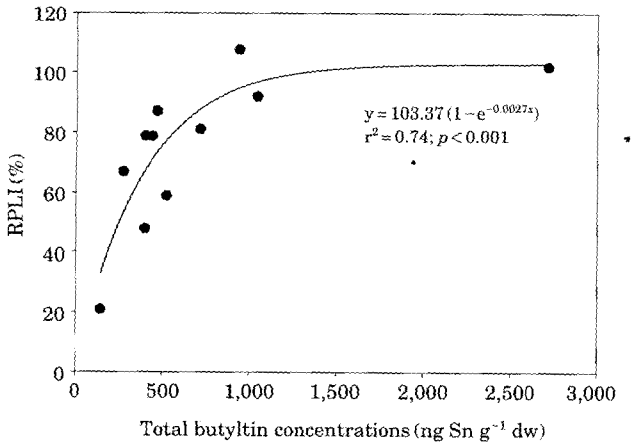

Fig. 4. Relationship between relative penis length index (RPLI) and concentrations of total butyltin (sum of mono-, di- and tributyltin) compounds in rock shell (*Thais clavigera*) collected from Gwangyang Bay.

are higher (81.2~107.9%) than those in the outer bay (20.9~78.9%) except for Site B13, (102.3%) where Gukdong Harbor is located (Fig. 1). This high frequency of imposex and its wide distribution in Gwangyang Bay indicate that organotin contamination is still enough to cause imposex in the bay, although TBT levels in sediment of the bay among the least in the bays in Korea. Only 47 and 56 ng g^{-1} TBT residue levels in the rock shell at Sites B15 and B14, respectively, were enough to induce imposex up to RPLI of 66.8%. Although the sediment TBT concentrations are in a decreasing trend in Gwangyang Bay for the past several years, it is not enough to protect rock shell from imposex casused by TBT contamination in the bay. Thus, long-term monitoring is required to assess effects of TBT on sensitive marine organisms in the bay.

4. Relationship between degree of imposex and butyltin body residues

All the rock shell samples contained detectable TBT compound (Table 4). The concentrations of TBT and total butyltin in rock shell ranged from 47 to 236 ng g^{-1} and from 144 to 2,717 ng g^{-1}, respectively. It is known that TBT is more easily degraded to DBT and MBT in rock shell than in oyster (Shim *et al.* 1998). Degradation products should be taken into account together with TBT for comparison of distributions and body residue analysis. Relatively high butyltin concentrations in rock shell were detected at Site B13 (2,717 ng g^{-1}) and B6 (1,040 ng g^{-1}), which are followed by Sites B7 and B5 located in the eastern part of the bay. The lowest total butyltin levels were found at Sites B14 and B15.

Relationship between RPLI and total butyltin concentration in rock shell is shown in Figure 4. The total butyltin body residues and degree of imposex in rock shell demonstrated a significant correlation ($r^2 = 0.74$; $p < 0.001$). These data provide supporting evidence for the dose-response relationship of TBT in the induction of imposex in rock shell. A curve of exponential rise to

maximum fitted the best in the case as same the previous study along the coast of Korea (Shim *et al.* 2000). In addition, the concentrations of total butyltin that caused half of RPL_{MAX}, estimated from the models, was 389 ng g^{-1}.

The degree of imposex appears to be closely related to boating activities, suggesting that antifouling paint applied to ship hulls was a major source of organotin contamination. In addition, many scattered small wharves and the fishing vessel traffic may also be sources leading to the wide distribution of imposex to the remote places from harbors and other shipping facilities.

ACKNOLEDGEMENT

This study is a part of project entilited "A study on management model for environmental pollution of special management area", which is supported by KORDI (project no. PE836-00).

REFERENCES

Alzieu C. 1986. TBT detrimental effects on oyster culture in France-evolution since antifouling paint regulation. In: Proceedings of the Oceans '86 International Organotin Symposium 4:1130-1134.

Bryan GW, PE Gibbs, LG Hummerstone and GR Burt. 1986. The decline of the gastropod, *Nucella lapillus*, around South-West England: evidence for the effect of tributyltin from antifouling paints. J. Mar. Biol. Assoc. U.K. 66:611-640.

Dowson PH, JM Bubb and JN Lester. 1992. Organotin distribution in sediments and waters of selected east coast estuaries in the U.K.. Mar. Pollut. Bull. 24:492-498.

Fent K. 1996. Ecotoxicology of organotin compounds. Crit. Rev. Toxicol. 26:1-117.

Gibbs BE, GW Bryan, PL Pascoe and GR Burt. 1987. The use of the dog-whelk, *Nucella lapillus*, as an indicator of tributyltin (TBT) contamination. J. Mar. Biol. Assoc. U.K. 67:507-523.

Horiguchi T, H Shiraishi, S Shimizu and M Morita. 1997. Effects of triphenyltin chloride and five other organotin compounds on the development of imposex in the rock shell, *Thais clavigera*. Environ. Pollut. 95:85-91.

Horiguchi T, H Shiraishi, M Shimizu and M Morita. 1994. Imposex and organotin compounds in *Thais clavigera* and *T. bronni* in Japan. J. Mar. Biol. Assoc. U.K. 74: 651-669.

Kan-atireklap S, S Tanab, J Sanguansin, S Tabucanon and M Hungspreugs. 1997. Contamination of butyltin compounds and organochlorine residues in green mussel (*Perna viridis*, L.) from Thailand coastal waters. Environ. Pollut. 97:79-89.

Kannan K, S Tanabe, H Iwata and R Tatsukawa. 1995. Butyltins in muscle and liver of fish collected from certain Asian and Oceanian countries. Environ. Pollut. 90: 279-290.

Ministry of Maritime Affairs and Fisheries (MOMAF). 1997. Annual statistics of fisheries and maritime. MOMAF, Seoul, Korea. 620pp. (in Korean)

Ministry of Maritime Affairs and Fisheries (MOMAF). 1998. Studies on TBT contamination in marine environment of Korea. MOMAF, Seoul, Korea. 211pp. (in Korean)

Moens L, T de Smaele, R Dams, P van den Broeck and P Sandra. 1997. Sensitive, simultaneous determination of organomercury, -lead, and -tin compounds with headspace solid phase microextraction capillary gas chromatography combined with inductively coupled plasma mass spectrometry. Anal. Chem. 69:1604-1611.

de Mora SJ, NG King and MC Miller. 1989. Tributyltin and total tin in marine sediments: profiles and the apparent rate of TBT degradation. Environ. Technol. Lett. 10:901-908.

Oehlmann J, P Fioroni, E Stroben and B Markert. 1996. Tributyltin (TBT) effects on *Ocinebrina aciculata* (Gastropod: Muricidae): imposex development, sterilization, sex change and population decline. Sci. Total Environ. 188:205-223.

Rodriguez I, S Mounicou, R. Lobinski, V. Sidelnikov, Y Patrushev and M Yamanaka. 1999. Species-selective analysis by microcolumn multicapillary gas chromatography with inductively coupled plasma mass spectrometric detection. Anal. Chem. 71:4534-4543.

Shim WJ, JR Oh, SH Kahng, JH Shi and SH Lee. 1998. Accumulation of tributyl- and triphenyltin compounds in Pacific oyster, *Crassostrea gigas*, from the Chinhae Bay System, Korea. Arch. Environ. Contam. Toxicol. 35:41-47.

Shim WJ, JR Oh, SH Kahng, JH Shim and SH Lee. 1999. Horizontal distribution of butyltins in surface sediments from an enclosed bay system, Korea. Environ. Pollut. 106:351-357.

Shim WJ. 2000. A study on the environmental chemistry and toxicology of organotins in the marine environment

of Korea. Ph.D thesis, Seoul National Universitiy, Seoul, Korea. 263pp.

Shim WJ, SH Kahng, SH Hong, NS Kim, SK Kim and JH Shim. 2000. Imposex in the rock shell, *Thais clavigera*, as evidence of organotin contamination in the marine environment of Korea. Mar. Environ. Res. 49:435-451.

Shim WJ, SH Hong, UH Yim, NS Kim and JR OH. 2002. Horizontal and vertical distribution of butyltin compounds in sediments from shipyards in Korea. Arch. Environ. Contam. Toxicol. 43:277-283.

Shim WJ, JK Jeon, SH Hong, NS Kim, UH Yim, JR Oh and YB Shin. 2003. Accumulation of tributyltin in olive flounder, *Paralichthys olivaceus*: its effect on hepatic cytochrome P450. Arch. Environ. Contam. Toxicol. 44: 390-397.

Stroben E, J Oehlmann and P Fioroni. 1992. *Hinia recticulata* and *Nucella lapillus*: Comparison of two gastropod tributyltin bioindicator. Ma. Biol. 114:289-296.

Sudaryanto A, S Takahashi, I Monrith, A Ismail, M Muchtar, J Zheng, BJ Richardson, A Subramanian, M Prudente, ND Hue and S Tanabe. 2002. Asia-Pacific mussel watch: monitoring of butyltin contamination in coastal waters of Asian developing countries. Environ. Toxicol. Chem. 21:2119-2130

Thompson JA, MG Sheffer, RC Pierce, YK Chau, JJ Cooney, WP Cullen and RJ Maguire. 1985. Organotin compounds in the aquatic environment: Scientific criteria for assessing their effects on environmental quality. National Research Council Canada. 284pp.

Tolosa I, L Merlini, N de Bertrand, JM Bayona and J Albaiges. 1992. Occurrence and fate of tributyl- and triphenyltin compounds in western Mediterranean coastal enclosures. Eviron. Toxicol. Chem. 11:145-155.

WHO. 1990. Tributyltin compounds, World Health Organization, Geneva. 273pp.

Yonezawa Y, K Nakata, Y Miyakozawa, A Ochi, T Kowata, H Fukawa, Y Sato, S Masunaga and Y Urushigawa. 1993. Distribution of butyltins in the surface sediement of Ise Bay, Japan. Environ. Toxicol. Chem. 12:1175-1184.

Manuscript Received: October 17, 2003
Revision Accepted: January 28, 2004
Responsible Editorial Member: Gi Hoon Song
(Korea Naval Academy)