

Occurrence of butyltin compounds in marine environment of Gwangyang Bay, Korea

Hee-Gu Choi*, Sang-Soo Kim, Hyo-Bang Moon and Bon-Kyu Gu

Marine Environment Management Division, National Fisheries Research & Development Institute, Busan 612-902, Korea

(Manuscript received 30 May, 2002 ; accepted 27 August, 2002)

Butyltin (BT) compounds were measured in seawater, sediment, sediment core, settling solids, and plankton from Gwangyang Bay in 2001. Tributyltin (TBT) was detected in seawater from 1 out of the 7 seawater sampling sites and in sediment from 18 out of the 35 sediment sampling sites. The highest concentration of TBT was found in the sediment from the site near Yeosu Harbor (53 ng Sn/g dry wt), acting as the point source for TBT. The mean concentrations of TBT were in the order of plankton > settling solids > sediment. The degradation indexes ($[(DBT)+[MBT])/[TBT]$) for the plankton were less than 1, indicating the possibility of recent inputs of TBT. The indexes for the sediment and settling solids ranged from 1.14 to 8.73. The composition of the BT compounds found in the settling solids was similar to that found in the sediment. The vertical profile of the total BT compounds in the sediment was characterized by an abrupt decline from the surface. However, no butyltin compounds appeared below a depth of 10 cm, corresponding to the 1980s. Accordingly, the current results demonstrated that the levels of all butyltin species in the environment of Gwangyang Bay were relatively lower than those in other polluted coastal areas. The vertical profile also suggested a fairly recent history for the down-core.

Key words : Butyltin, seawater, sediment, sediment core, settling solid, plankton, TBT, degradation indexes

1. Introduction

Tributyltin (TBT), which is the main active component in organotin-based antifouling paints, is possibly one of the most toxic substances that is intentionally introduced into marine environments¹⁾. Because of its ability to cause toxicity at very low water concentrations, the use of TBT has been restricted since the 1980s in many countries^{2,3)}. Nonetheless, TBT remains an important pollutant in areas with active shipyards and commercial shipping traffic. Recent studies have indicated the widespread occurrence and accu-

mulation of butyltin compounds in the sediment and organisms^{4~6)}, even though TBT levels in seawater have declined since regulation^{7,8)}. In Korea, the use of TBT-based paints at fishery facilities and on small vessels was unregulated until 1999, despite concerns over the impacts of TBT. A few studies on antifoulant concentrations in the Korean marine environment are available^{9~13)}. However, these studies only focused on seawater, sediment, and organisms. Gwangyang Bay, surrounded by the POSCO, Yeochon, and Yulchon industrial complexes, is one of the most polluted coastal areas in Korea. Accordingly, the objectives of the current study were to estimate the occurrence of butyltin (BT) compounds in various environmental compartments of Gwangyang Bay, including the settling solids, as the intermediate media to sediment, plankton, as the primary source in the food chain, seawater, and sediment, and to

Corresponding author ; Hee-Gu Choi, Marine Environment Management Division, National Fisheries Research & Development Institute, Gijang-gun, Gijang-up, Busan 612-902, Korea
Phone : +82-51-720-2530
E-mail : hgchoi@nfrdi.re.kr

explore the historic input of BT compounds in the bay through a vertical profile of the sediment.

2. Materials and Methods

2.1 Sampling

Samples of seawater, sediment, sediment core, settling solids, and plankton were all collected from Gwangyang Bay in 2001 (Fig. 1). The seawater samples were collected using a bucket and taken from just beneath the surface at 7 sites (stations 9, 11, 14, 21, 24, 27, and 29) in April. The surface sediments (top 2 cm) were taken using a box sampler at 35 stations in April. The sediment core sample was obtained by divers from a site near the Gwangyang Container Terminal (station A) using an acrylic tube (8 cm diameter and 150 cm

long) in April. The settling solids were also sampled using nine replicate sediment trap arrays that were moored 3 m above the bottom sediment for 48 hours at 2 sites (stations B and C) in September. Each array consisted of four acrylic cylinders 7 cm in diameter and 64 cm long. The plankton samples were taken by towing horizontally for 12 hours using a 20 μm mesh net for phytoplankton and 330 μm mesh net for zooplankton at the mouth of the bay in September 2001.

2.2 Sample preparation

The samples were stored in a cooler box using ice or dry ice and transported to the laboratory immediately. The seawater was analyzed in within a week, while the other samples were stored at -20°C and later freeze-dried.

2.3. Chemical analysis

The analytical procedure was performed after combining and modifying the procedures of Wade et al.¹⁴⁾ and Harino et al.¹⁵⁾. Briefly, 1 L of the water samples was extracted twice with 0.1 % tropolone-methylene chloride (DCM) under the addition of conc-HCl and 10 g NaCl. The DCM layer was concentrated to almost dryness and exchanged into *n*-hexane. The extract was propylated with *n*-propyl magnesium bromide and then extracted using *n*-hexane. After clean-up with a florisil column, the *n*-hexane layer was determined using a gas chromatograph (Hewlett-Packard 6890) with a flame photometric detector and 610 nm cut-off interference filter. A capillary column HP 5 (5 % phenyl methyl siloxane, 30 m long, 0.25 mm internal diameter, 0.25 μm film thickness) was used. The temperature program was set as follows: initial at 80°C , $80\sim 160^\circ\text{C}$ at $15^\circ\text{C}/\text{min}$, $160\sim 200^\circ\text{C}$ at $5^\circ\text{C}/\text{min}$, 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 the carrier and make-up gas at flow rates of 1.4 ml/min and 18.6 ml/min, respectively. Hydrogen and air gases were passed at 150 ml/min and 100 ml/min, respectively. The freeze-dried samples of the sediment, settling solids, and plankton were extracted with 0.1 % tropolone-DCM after the addition of 1 N HCl. For the sediment, the organic

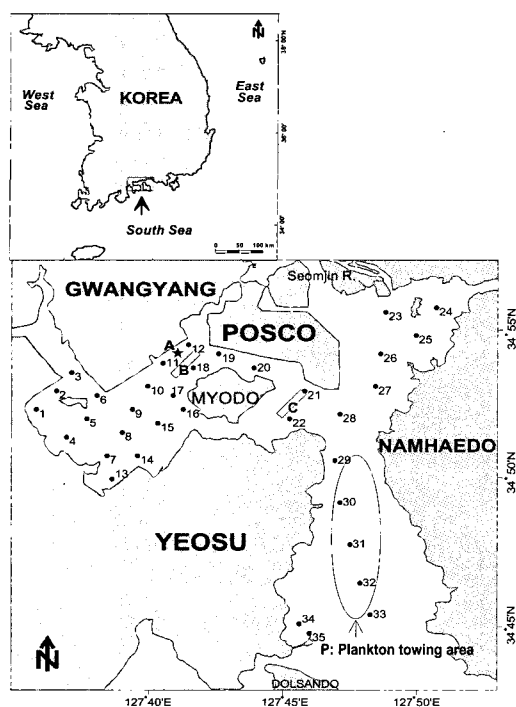


Fig. 1. Map showing the location of sampling stations. Seawater samples were collected from Sts. 9, 11, 14, 21, 24, 27 and 29. Sediment samples were collected from Sts. 1-35. Sediment core was collected from St. A. Settling solids were collected from Sts. B and C. Plankton samples were collected from the mouth of bay (St. P).

layers were washed with 3.3% tetrabutyltinammonium hydrogensulphate and 16% sodium sulfide to remove any co-extracted inorganic sulfur-containing species before the propylation. After concentrating the organic layer, the subsequent procedures were identical with those for the water samples. Tripentyltin chloride was spiked into all samples before extraction as a surrogate standard. The extracts eluted with *n*-hexane were concentrated and tetrabutyltin (internal standard) was added to estimate the recoveries. The values were given in ng Sn/L for seawater and ng Sn/g dry wt for the sediment, settling solids, and plankton. The quality assurance and quality control procedures included internal standards, procedural blanks, and an analysis of reference materials (NIES No 11; Sea bass, Japan and PACS-2; sediment, Canada). The recoveries with the reference materials were more than 90%. The detection limits for each butyltin compound were around 2 ng Sn/L in the seawater and around 3 ng Sn/g dry wt in the sediment.

3. Results and Discussion

3.1 Levels of butyltin compounds

The concentrations of butyltin compounds in the different environment compartments are shown in Table 1.

Seawater: Tributyltin was only detected in seawater from one (station 9: 7 ng Sn/L) of the 7 stations used to sample the seawater in Gwangyang Bay. Generally, the TBT found in an aquatic environment is the result of leaching from antifouling paints, although other sources have also been identified¹⁶. However, in the current study, the source was unclear as no TBT was detected at the sites near potential TBT sources, such as the Gwangyang Container Terminal. Yet, the TBT level detected at station 9 was still below the no-observed effect level (NOEL: 8 ng Sn/L for Pacific oysters¹⁷). As such, the current results were similar to previous data measured in 1996¹³. Accordingly, based on this information, the butyltin contamination status of Gwangyang Bay appeared to be similar to that in unpolluted areas.

Sediment: Tributyltin was detected in the

Table 1. Butyltin concentrations (seawater: ng Sn/L, other samples: ng Sn/g dry wt) in the aquatic environment of Gwangyang Bay

Sample	Site #	MBT	DBT	TBT	∑ BTs	
Seawater	9	nd*	nd	7	7	
	11	nd	nd	nd	nd	
	14	14	nd	nd	nd	
	21	nd	nd	nd	nd	
	24	nd	nd	nd	nd	
	27	nd	nd	nd	nd	
	29	nd	nd	nd	nd	
	Sediment	1	nd	nd	nd	nd
		2	26	nd	12	38
		3	28	nd	nd	28
		4	19	nd	6	25
		5	29	nd	nd	29
		6	21	11	7	38
		7	38	7	nd	45
		8	nd	nd	nd	nd
		9	48	9	11	68
		10	37	9	13	59
		11	43	nd	9	52
		12	34	9	14	57
13		25	nd	nd	25	
14		35	nd	nd	35	
15		48	nd	nd	48	
16		23	nd	nd	23	
17		35	nd	10	44	
18	39	9	13	61		
19	22	15	nd	37		
20	39	10	8	57		
21	nd	nd	nd	nd		
22	39	16	15	69		
23	nd	nd	nd	nd		
24	56	35	39	131		
25	26	nd	nd	26		
26	nd	nd	nd	nd		
27	31	8	12	51		
28	49	nd	nd	49		
29	nd	nd	9	9		
30	nd	19	nd	19		
31	22	15	nd	37		
32	30	5	4	40		
33	11	4	7	21		
34	39	22	53	114		
35	53	28	31	112		
Sediment core A	(0-2 cm)	23	17	48	88	
	(2-4 cm)	10	8	9	28	
	(4-6 cm)	19	8	9	36	
	(6-8 cm)	17	2	nd	19	
	(8-10 cm)	11	nd	nd	11	
Settling solid	B	62	14	19	94	
	C	44	11	27	82	
Phytoplankton	P	228	115	388	730	
Zooplankton	P	57	35	140	232	

* Not detected

sediment from 18 out of 35 stations used to sample the sediment in Gwangyang Bay. The concentrations of TBT ranged from 4 to 53 ng Sn/g dry wt, indicating the influence of the sampling site on the butyltin content. The highest concentration of TBT was found at station 34 adjacent to Yeosu Harbour, acting as the point source for TBT. When compared to the PADDA SL (Puget Sound Dredge Disposal Analysis Screening Level) of 29.8 ng Sn/g dry wt for TBT¹⁸⁾, only three samples (stations 24, 34, and 35) exceeded the PSDDA SL. In fact, the concentrations of TBT in the sediment from Gwangyang Bay were very low in comparison to the levels detected in sediments from other coastal areas (Chinhae Bay: 4~382 ng Sn/g dry wt¹¹⁾; Western Mediterranean: 51~7,642 ng Sn/g dry wt¹⁹⁾; Otsuchi Bay: 4~261 ng Sn/g dry wt²⁰⁾). The current results were 1 order of magnitude higher than previous data measured in the same area¹³⁾. In Korea, the use of TBT-based paints at fishery facilities and on small vessels only became regulated in 1999. Therefore, the low levels of TBT in the bay may not have been due to the effect of the regulation. A more likely explanation is physical effects, such as sediment dredging, seawater diffusion, or advective sediment transport. In fact, sediment dredging occurred in the 1980s, plus the tidal current in Gwangyang Bay is relatively strong²¹⁾.

Sediment core: The sediment core exhibited a rapid decline in butyltin content with an increasing depth, showing the highest TBT level within the surface 0~2 cm sediment. This pattern was consistent with a few other studies^{10,22)}. No butyltin compounds were detected at a depth of 7 cm for TBT, 9 cm for DBT (dibutyltin), and 11 cm for MBT (monobutyltin). According to a sedimentation rate of 0.68 cm/yr, as reported by Kang and Lee²³⁾, the time periods for those depths corresponded to 1989, 1986, and 1983, respectively. Therefore, it would appear that sediments prior to the early 1980s did not contain any detectable levels of BT compounds, even though organotin-based anti-fouling paints were first used in the 1960s²⁴⁾. Interestingly, this period coincides with the period when sediment dredging was carried out in Gwangyang Bay. Consequently, non-detection of TBT may have resulted from the removal of

contaminated bottom sediment by dredging.

Settling solids: The concentrations of TBT in the settling solids ranged from 19 ng Sn/g dry wt (station B near the Gwangyang Container Terminal) to 27 ng Sn/g dry wt (station C close to POSCO). The TBT concentrations in the settling solids were comparable to those in the sediment, yet lower than those in the plankton. According to Ko and Baker's study on the source of particles in surface and bottom waters²⁵⁾, more than 65 % of the suspended particles in surface water are plankton, while more than 50 % of the suspended particles in bottom water are resuspended sediment. In the current study, the sediment trap was deployed above 3 m from the seafloor. Therefore, when taking into consideration the effect of the composition of the suspended particles on the settling solids, resuspended sediment was likely to play a significant role in supplying particles to the water column of Gwangyang Bay.

Plankton: The concentration of butyltin compounds in the plankton was high in comparison to the levels in the other samples from Gwangyang Bay. The concentration of TBT was 388 ng Sn/g dry wt in the phytoplankton and 140 ng Sn/g dry wt in the zooplankton. Harino et al. also reported high levels of TBT in plankton in different aquatic environments at Otsuchi Bay²⁰⁾ and the Port of Osaka²⁶⁾. More recent studies have shown that BT compounds have a low affinity to lipids and less biomagnification capacity in prey-predator relationships than organochlorines^{16,27~29)}. In the current study, no considerable biomagnification was observed between the BT compounds in the phytoplankton and the zooplankton. The concentration of TBT in the phytoplankton was 2.8 times higher than that in the zooplankton. The TBT levels found in the plankton from Gwangyang Bay were similar to those found in the plankton from Eckwarderhorne in the North Sea³⁰⁾, yet 10 orders of magnitude lower than the 4,000 ng Sn/g dry wt measured at a site near a shipyard in Otsuchi Bay²⁵⁾.

3.2. Composition of TBT and its degradation products

The degradation of butyltins has been reported

to proceed by successive debutylation reactions to inorganic tin^{31~33}). TBT degradation can result from various mechanisms, such as photolysis and biodegradation. Photolysis by sunlight is the fastest route of degradation in water, while biological action is the most important route of degradation in sediment³¹. It is known that TBT degradation rates in sediment (half-lives: 365~775 days³²) are slower those in a water column (half-lives: 6~19 days³³). Therefore, it is useful to calculate the butyltin degradation index (BDI) expressed as the ratio between TBT and its degradation products (DBT and MBT) to identify the recent contamination by butyltin compounds¹⁹. The current results are presented in Fig. 2. The lowest BDI in sediment was observed at the highest polluted station 34 (1.14), while the highest BDI was found at station 32 located in the mouth of the bay (8.73). The other stations where sediment was collected exhibited a BDI between 2.10 and 5.10. The difference in the BDI among the various sediment samples can be explained by the effect of several factors, including the type and rate of degradation reaction, sediment characteristics, butyltin source signature, and processes, such as resuspension, adsorption, and desorption. The indexes for the settling solids were similar to those for the sediment, ranging from 2.00 to 4.01. The BDI values for the plankton were less than 1. A high TBT/total butyltin concentration ratio indicates only minimal TBT degradation and/or recent input of TBT to the ecosystem³⁴. As such, the low BDI for the plankton due to a high contribution of TBT may be evidence of recent TBT inputs. Harino et al.⁶ also reported that TBT was the major contributor of butyltins in plankton samples. Meanwhile, the intermediate values for the sediment and settling solids indicated a mild degradation process over parent compounds. However, it was difficult to identify how recent the butyltins inputs were. As regards the composition of the butyltin compounds in the samples, the distribution patterns of TBT and its breakdown products in the settling solids were similar to those in the sediment, yet different from those in the plankton (Fig. 3). The components of settling particles are similar to those of suspended solids and composed of a mixture of algal debris and resuspended sediment with various degrees of

mineralization³⁵). Among the different particles, resuspended sediment contributes from 50 % to 100 % of the total suspended particles in bottom water²⁵). Consequently, it can be assumed that resuspended sediment played a larger role in the settling flux due to the location of the sediment traps deployed 3 m above the bottom sediment. The vertical profile of the sediment core revealed that the contribution of TBT decreased with a concomitant increase in MBT according to the depth (Fig. 4). A few other studies have revealed different results for TBT degradation relative to depth. De Mora et al.³⁶ observed an exponential decrease in TBT levels with depth, whereas Steward and de Mora³⁷ found no clear pattern with depth. The probable causes would appear to be the biological activities of benthic organisms along with physical mixing³⁸). In the case of Gwangyang

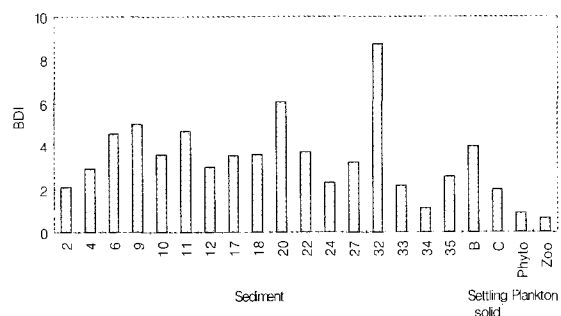


Fig. 2. Butyltin degradation indexes (BDI) in sediment, settling solid and plankton. BDI is calculated as follows: $BDI = \frac{[MBT] + [DBT]}{[TBT]}$.

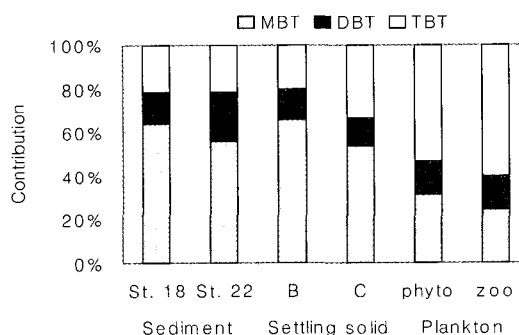


Fig. 3. Comparison of contribution of TBT and its metabolites among environment compartments.

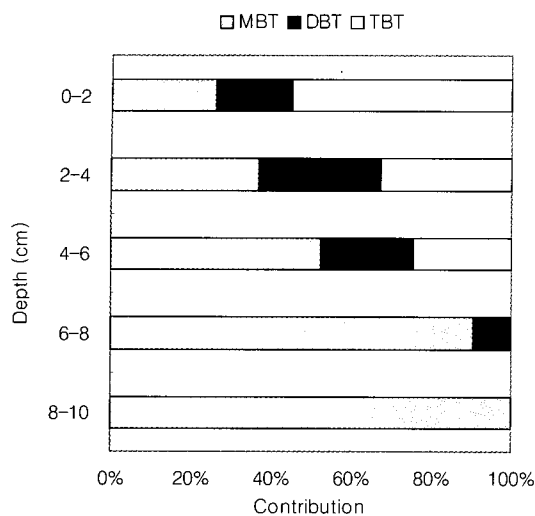


Fig. 4. Vertical profiles on the contribution of TBT and its degradation products in sediment core.

Bay, the most likely explanation for the composition of butyltin compounds in the sediment core was physical factors related to the sediment dredging that occurred in the 1980s and a strong tidal current.

4. Conclusions

The levels of tributyltin (TBT) found in the aquatic environment of Gwanyang Bay were relatively low compared to those in other polluted coastal areas. Tributyltin was only detected in seawater from one site near Yeosu Harbor. The TBT concentrations in the other samples were within a range of 4~53 ng Sn/g dry wt for the sediment, 19~27 ng Sn/g dry wt for the settling solids, and 140~388 ng Sn/g dry wt for the plankton. TBT was detected in sediment from 18 out of 35 test sites and the highest concentration was from the site near Yeosu Harbor. The mean concentrations of butyltin compounds in the environment compartments decreased as follows: plankton > settling solids > sediment. As regards the composition of the BT compounds, the contributions of TBT differed according to the environment media. In the plankton, the dominant compound was TBT, while in the settling solids and sediment it was monobutyltin (MBT). The down-core trend exhibited the maximum TBT concentration at the surface, which then decreased

with depth. However, no butyltin compounds were detected below a depth of 10 cm, corresponding to the 1980s.

References

- [1] Mee, L.D. and S.W. Fowler, 1991, Organotin biocides in the marine environment: A managed transient Mar. Environ. Res., 32, 89~111.
- [2] Chau, Y.K., R.J. Maguire, M. Brown, F. Yang and S.P. Batchelor, 1997, Occurrence of organotin compounds in the Canadian aquatic environment five years after the regulation of antifouling uses of tributyltin. Water Qual. Res. J. Canada, 32, 453~521.
- [3] Meador, J.P., 1997, Comparative toxicokinetics of tributyltin in five marine species and its utility in predicting bioaccumulation and acute toxicity. Aqua. Toxicol., 37, 307~326.
- [4] Fent, K., 1999, Ecotoxicology of organotin compounds. Critical Review of Toxicology, 26, 1~117.
- [5] Kannan, K. and J. Falandysz, 1997, Butyltin residues in sediment, fish, fish-eating birds, harbour porpoise and human tissues from the Polish coast of the Baltic Sea. Mar. Pollut. Bull., 34, 203~207.
- [6] Harino, H., M. Fukushima and S. Kawai, 1999, Temporal trends of organotin compounds in the aquatic environment of the Port of Osaka, Japan. Environ. Pollut., 105, 1~7.
- [7] Alzieu, C., J. Sanjuan, J.P. Deltriel and M. Borel, 1986, Tin contamination in Arcachon bay: effects on oyster shell anomalies. Mar. Pollut. Bull., 17, 494~498.
- [8] Evans, S.M., A.C. Birchenough and H. Fletcher, 2000, The value and validity of community-based research: TBT contamination of the North Sea. Mar. Pollut. Bull., 40, 220~225.
- [9] Choi, H.G., P.J. Kim and W.C. Lee, 1997, Butyltin compounds concentrations in Masan Bay. J. Kor. Fish. Soc., 30, 923~928.
- [10] Hwang, H. M., J.R. Oh, S.H. Kahng and K.W. Lee, 1999, Tributyltin compounds in mussels, oysters and sediments of Chinhae Bay, Korea. Mar. Environ. Res., 47, 61~70.

- [11] Shim, W.J., J.R. Oh., S.H. Kahng, J.H. Shim and S.H. Lee, 1999, Horizontal distribution of butyltins in surface sediments from an enclosed bay system, Korea. *Environ. Pollut.*, 106, 351~357.
- [12] Hong, H.K., S. Takahashi, B.Y. Min and S. Tanabe, 2002, Butyltin residues in blue mussels (*Mytilus edulis*) and arkshells (*Scapharca broughtonii*) collected from Korean coastal waters. *Environ. Pollut.*, 117, 475~486.
- [13] Kim, G.Y., 1997. Evaluation of butyltin compounds and its distribution among seawater, sediments and biota from Kwangyang Bay. Master thesis. Pukyong National Univ.
- [14] Wade, T.L., B. Garcia-Ramero and J.M. Brooks, 1988, Tributyltin contamination in bivalves from United States coastal estuaries. *Environ. Sci. Technol.*, 22, 1488~1493.
- [15] Harino, H., M. Fukushima and M. Tanaka, 1992, Simultaneous determination of butyltin and phenyltin compounds in the aquatic environment by gas chromatography. *Anal. Chim. Acta*, 264, 91~96.
- [16] Takahashi, S., S. Tanabe, I. Takeuchi and N. Miyazaki, 1999. Distribution and specific bioaccumulation of butyltin compounds in a marine ecosystem. *Arch. Environ. Contam. Toxicol.*, 37, 50~61.
- [17] Alzieu, C., 1991, Environmental problems caused by TBT in France: assessment, regulations, prospects. *Mar. Environ. Res.*, 32, 7~17.
- [18] Elgethun, K., C. Neumann and P. Blake, 2000, Butyltins in shellfish, finfish, water and sediment from the Coos Bay estuary (Oregon, USA). *Chemosphere*, 41, 953~964.
- [19] Díez, S., M. Ábalos and J.M. Bayona, 2002, Organotin contamination in sediments from the Western Mediterranean enclosures following 10 years of TBT regulation. *Wat. Res.*, 36, 905~918.
- [20] Harino, H., M. Fukushima, Y. Yamamoto, S. Kawai and N. Miyazaki, 1998, Contamination of butyltin and phenyltin compounds in the marine environment of Otsuchi Bay, Japan. *Environ. Pollut.*, 101, 209~214.
- [21] Lee, S. U., 1994, Korean harbour hydrology. *Jipmundang*, 117~120.
- [22] Venkatesan, M.I., V. Chalaux, J.M. Bayona and E. Zeng, 1998, Butyltins in sediments from Santa monica and San Pedro basins, California. *Environ. Pollut.*, 99, 263~269.
- [23] Kang, H.W. and K.W. Lee, 1996, Trace metal concentrations and sedimentation rates of Kwangyang, Masan and Ulsan Bay sediments. *J. Korean Soc. Wat. Qual.*, 12, 455~461.
- [24] Stewart, C., A.J. Thompson, 1994, Extensive butyltin contamination in southwestern coastal British Columbia, Canada. *Mar. Pollut. Bull.*, 28, 601~606.
- [25] Ko, F.C. and J.E. Baker, 1995. Partitioning of hydrophobic organic contaminants to resuspended sediments and plankton in the mesohaline Chesapeake Bay. *Mar. Chem.*, 49, 171~188.
- [26] Harino, H., M. Fukushima and S. Kawai, 1999, Temporal trends of organotin compounds in the aquatic environment of the Port of Osaka, Japan. *Environ. Pollut.*, 105, 1~7.
- [27] Bryan, G.W., and P.E. Gibbs, 1991. Impact of low concentrations of tributyltin (TBT) on marine organism: a review. In: Newman MC. McIntosh AW (eds) *Metal ecotoxicology: concepts and applications*. Lewis Publishers, Ann Arbor MI, 323~335.
- [28] Yamada, H. and K. Takayanagi, 1992. Bioconcentration and elimination of bis (tributyltin) oxide (TBTO) and triphenyltin chloride (TPTC) in several marine fish species. *Water Res.*, 12, 1589~1595.
- [29] Kannan, K., S. Tanabe, R. Tatsukawa and R.J. Williams, 1995. Butyltin residues in fish from Australia, Papua New Guinea and the Solomon Islands. *Intern J. Environ. Anal. Chem.*, 61, 263~273.
- [30] Shawky, S. and H. Emons, 1998, Distribution pattern of organotin compounds at different trophic levels of aquatic ecosystems. *Chemosphere*, 36, 523~535.
- [31] Hoch, M., 2001. Organotin compounds in the environment - an overview. *Appl. Geochem.*, 16, 719~743.
- [32] Dowon, P.H., J.K. Bubb, T.P. Williams and J.N. Lester, 1993. Degradation of tributyltin in sediment in freshwater and estuarine marine sediments. *Wat. Sci. Technol.*, 28, 133

- ~ 137.
- [33] Seligman, P.F., A.O. Valkirs and R.F. Lee, 1986. Degradation of tributyltin in San diego Bay, California, Waters. Environ. Sci. Technol., 20, 1229~1235.
- [34] Gui-bin, J., Z. Qun-fang, L. Ji-yan and W. Di-jing, 2001, Occurrence of butyltin compounds in the waters of selected lakes, rivers and coastal environments from China. Environ. Pollut., 115, 81~87.
- [35] Koekmans, A.A., F. Gillissen, W. Makatita and Van den Berg, M., 1997, organic carbon normalization of PCB, PAH, and pesticide concentrations in suspended solids. Wat. Res., 31, 461~470.
- [36] de Mora, S.J., N.G. King and M.C. Miller, 1989, Tributyltin and total tin in marine sediments: profiles and the apparent rate of TBT degradation. Environ. Technol. Letters, 10, 901~908.
- [37] Stewart, C. and S.J. de Mora, 1992, Elevated tri (n-butyl) tin concentrations in shellfish and sediments from Suva Harbor, Fiji. App. Organo. Chem., 6, 507~512.
- [38] Page, D.S., C.C. Ozbal and M.E. Lanphear, 1996, Concentration of butyltin species in sediments associated with shipyard activity. Environ. Pollut., 91, 237~243.