

# Distribution of Organotin Compounds in Representative Coastal Areas from Japan: A Review

Hiroya HARINO<sup>1\*</sup>, Sayaka EGUCHI<sup>2</sup>, Yoshikazu YAMAMOTO<sup>2</sup>, Yuko KUROKAWA<sup>2</sup>, Shin'ichiro KAWAI<sup>2</sup>, Takaomi ARAI<sup>3</sup>, Madoka OHJI<sup>4</sup>, Shigeyuki YAMATO<sup>5</sup>, Naomasa KOBAYASHI<sup>6</sup> and Nobuyuki MIYAZAKI<sup>7</sup>

<sup>1</sup> Osaka City Institute of Public Health and Environmental Sciences, Tojo-cho 8–34, Tennoji-ku, Osaka, 543–0026 Japan

\* E-mail: j-harino@kawachi.zaq.ne.jp

<sup>2</sup> Department of Human Sciences, Kobe College, Okadayama 4–1, Nishinomiya, Hyogo 662–0827 Japan

<sup>3</sup> International Coastal Research Center, Ocean Research Institute, University of Tokyo, 106–1, Akahama 2-chome, Otsuchi-cho, Iwate 028–1102 Japan

<sup>4</sup> Institute of Symbiotic Science and Technology, Tokyo University of Agriculture and Technology, Fuchu, Tokyo 183–8509, Japan

<sup>5</sup> Seto Marine Biological Laboratory, Kyoto University, Shirahama, Nishimuro, Wakayama, 649–2211 Japan

<sup>6</sup> Professor Emeritus, Doshisha University, 26 Moroki-cho, Kamitakano, Saikyo-ku, Kyoto-shi, 606–0046

<sup>7</sup> Center for International Cooperation, Ocean Research Institute, The University of Tokyo, 1–15–1, Minamidai, Nakano, Tokyo, 164–8639, Japan

»» Received 5 November 2007; Accepted 17 December 2007

**Abstract**—The temporal trends of the organotin (OT) compounds in the coastal waters from Japan were investigated. Tributyltin (TBT) concentrations in water and biological samples decreased drastically in 1990 which regulation was enacted and after that, the decreasing of TBT concentration were not observed. No remarkable changes of TBT concentrations in sediment were observed in this period.

At the present day, mean concentration of TBT in water from Japan was  $0.011 \mu\text{g l}^{-1}$  and no remarkable differences of TBT concentrations among sampling sites were observed. On the other hand, TBT concentrations were high in sediment from Ofunato Bay, the Port of Osaka, Tanabe Bay and Hakata Bay with poor flushing and shallow water. TBT concentrations in Tanabe Bay were the highest among the surveyed coastal waters of Japan. OTs concentrations were high in mussels from Yamada Bay and Otsuchi Bay which have many fishing ports.

The vertical distribution in water column from Osaka Bay was discussed. TBT concentrations were constant during 0–20 m of water depth, which is due to vertical mixture of water. In the sediment cores, the concentration of TBT in the top 10 cm was higher than those in the other sediment core. It is estimated from the accumulation pattern and the deposition rate that TBT has been roughly used since the 1960.

Cross-media partitioning of TBT in aquatic environment were calculated from the concentrations in water, sediment and biological samples. The sediment-water partitions coefficient and the biota-water partitions of TBT were high. Considering cross-media partitioning and the half-lives of TBT in each compartment, it found that TBT contaminations in sediment have persisted for a long time. In order to recovery the OTs contamination, it is important to remove the sediment which was contaminated by OTs.

**Key words:** butyltin, phenyltin, water, sediment, biological sample

## Introduction

Antifouling products contain biocides which were developed to prevent the progressive built up of bacteria, macro algae, mussels, barnacles and other invertebrates in water surfaces. Organotin (OT) compounds are chemical substances which have been used as an effective antifouling biocide. However, it was reported that marine organisms such as dog-whelk (*Nucella lapillus*) and Pacific oyster (*Crassostrea gigas*) have been damaged by leaching of OTs into water col-

umn from coatings applied to boat hulls (Alizieu et al. 1986, Gibbs and Bryan 1986). Many studies on the current status of OTs contamination and toxicity of OTs for aquatic organism were carried out. As the result, in the developed countries such as England, USA, Canada etc., the use of butyltin (BT) compounds has regulated, for instance, establishment of environmental quality target, control of release rates from antifouling paint and ban on the use of Tributyltin (TBT)-containing antifouling paints on pleasure crafts less than 25 m length. Although the use of TBT based antifouling paints in developed countries has been restricted, coastal areas found

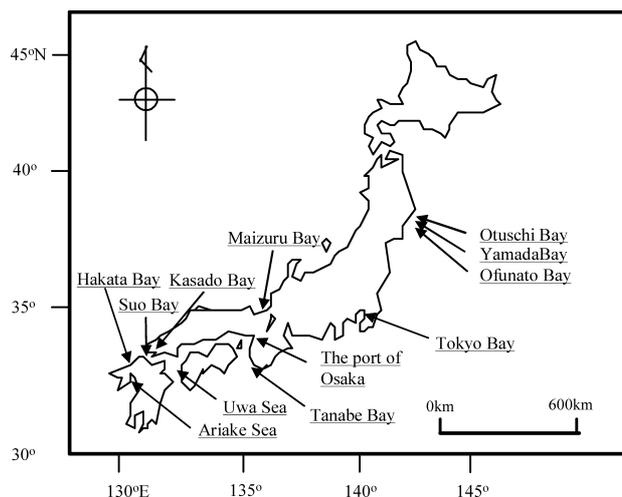
to be contaminated widely by OTs (Harino et al. 1998a, Chau et al. 1997). Furthermore, higher concentration of OTs was detected in developing countries which the regulation for OTs is not enacted (Harino et al. 2006, Midorikawa et al. 2004, Sudaryanto et al. 2002). As OTs contamination have still continued in the worldwide, the International Maritime Organization (IMO) adopted the International Convention on the Control of Harmful Antifouling Systems (AFS Convention) which will prohibit the use of OTs as active ingredients in antifouling systems on ships in January 2008.

In Japan, bis(tributyltin)oxide (TBTO) has been designated a Class 1 Specified Chemical Substance under “the Law Concerning the Examination and Regulation of Manufacture etc. of Chemical Substances” in 1989, and triphenyltin (TPT) compounds (7 chemicals) and TBT species excluding TBTO (13 chemicals) have been designated as Class 2 Specified Chemical Substances in 1990 under the same Law. It has been required to report the amounts of those compounds which are manufactured and imported to the Ministry of Economy, Trade and Industry by this law. There has been no such a report indicating the use of OTs in Japan, since 1996. In 1992, Environment Agency also set a guideline recommending that OTs concentration within the area of 100 m from source should be  $0.1 \mu\text{g/l}$  or less and other areas to be  $0.01 \mu\text{g/l}$  or less. In spite of these regulations, it is reported that the higher concentrations of OTs have still been detected in the coastal water in Japan (Harino et al. 1999).

Up to date, some papers concerning the current status of OTs in Japan were published and some issues regarding the OTs contamination were indicated. It needs to summarize the various issues in the coastal water of Japan before the worldwide inhibition of the usage of OTs. In this review, temporal trend of OTs, horizontal distribution of OTs contamination and cross-media partitioning of OTs in representative coastal waters of Japan are summarized, and the issues on the OT contamination which will continue in near future, are discussed.

### Temporal trend of organotin compounds

In the global view point, temporal trend of OTs in coastal waters of Japan is discussed by Ministry of the Environment (2004). Ministry of the Environment has surveyed the TBT and TPT compounds in 35 sites of coastal water in Japan for a long time. The concentrations of TBT and TPT in water and sediment decreased between 1990 and 2002. Environmental Agency has also surveyed the TBT and TPT in mussel, fish and bird between 1985 and 2002. Although TBT and TPT concentrations in these animals decreased in the start of the survey, concentrations of these chemical compounds were constant in recent years. Takeuchi et al. (2004) also investigated BTs in water samples from 18 coastal areas in Japan between 1997 and 1999, and they found that TBTO concentrations has not significantly declined during that pe-



**Figure 1.** The map of study area

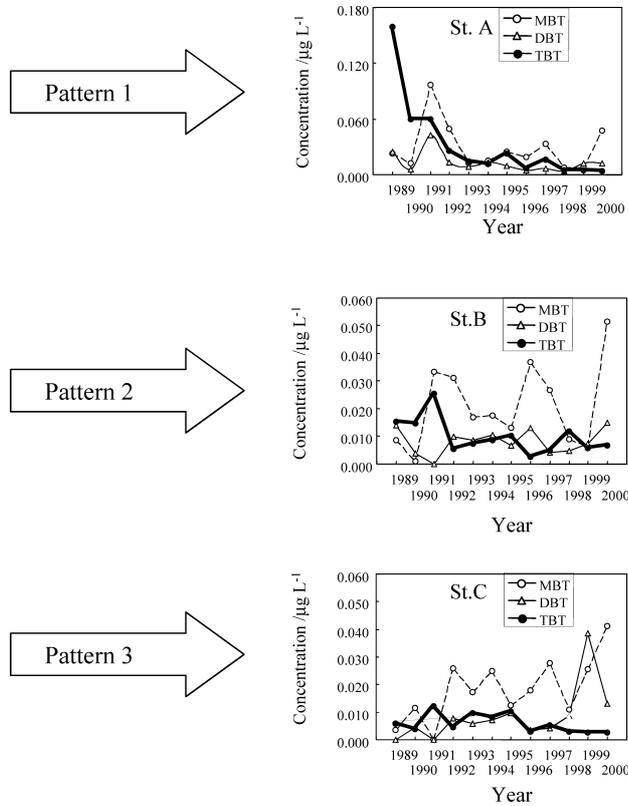
riod.

The temporal trend of OTs in some local areas was discussed in detail. Harino et al. (2004) has monitored BTs and phenyltin (PT)s in the Port of Osaka, an international trading port, where about 80,000 ships sail each year (Fig. 1). Surveys at eight stations in this area were undertaken to evaluate the temporal change of TBT level between 1989 and 2000. Three patterns of temporal distribution of TBT were apparent in water from the Port of Osaka. The first pattern was characterized by dramatic decrease of TBT concentration during 1989–1991 (Fig. 2 pattern 1). The stations which belong to this type were at marina and berthing facilities of Japanese-registered small-hull crafts with poor flushing. TBT concentrations decreased by 4- to 10- fold during 1989–1991 at these stations. Although TBT became dominant compound among BTs during 1989–1991, the ratios of combined monobutyltin (MBT) and dibutyltin (DBT) were higher than those of TBT in recent years. These results show that the input of TBT has decreased considerably by regulation in marina and berthing facilities of Japanese-registered small-hull crafts.

The second pattern is general trend in the water of the Port of Osaka, showing a slight reduction of TBT concentration during 1989–2000 (Fig. 2 pattern 2). The dominant species of BTs changed from TBT to MBT as degradation product of TBT in this period. A large number of ships from various countries sail and is moored at the Port of Osaka as an international trading port. Although the inputs of OTs from Japanese-registered vessels have decreased, TBT inputs from vessels from non regulated countries have still continued. The third pattern is characterized by constant levels of TBT between 1989 and 2000 (Fig. 2 pattern 3). MBT was the dominant among BTs between 1990 and 2000. The sampling site of pattern 3 was located in the Japanese ferry port, suggesting that BTs had not been used in Japanese ferry boats in 1989. PT compounds in water from the Port of Osaka were

detected sometimes during 1989–1990, and after that, PTs were not detected at any of the stations.

Figure 3 shows the temporal trend of BTs concentration in sediment at representative sampling sites in the Port of

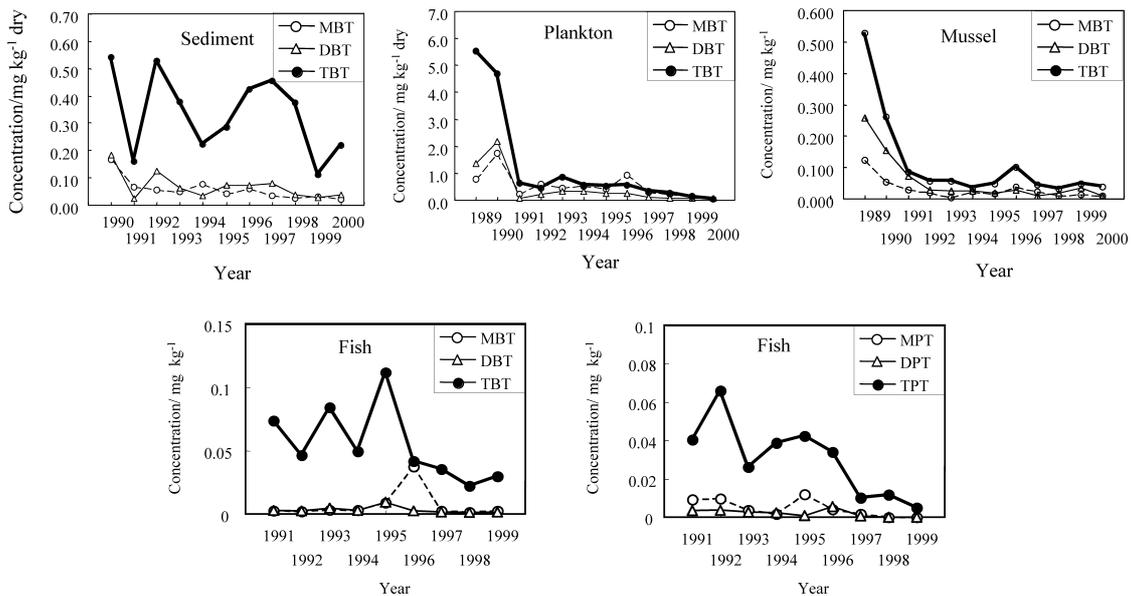


**Figure 2.** The temporal trend of BTs in water from the Port of Osaka. (Harino et al. 2004).  
 A: Marina, B: Most sites in the Port of Osaka, C: Japanese ferry port

Osaka. No change of TBT concentrations were observed between 1990 and 2000. The half-lives of TBT in sediment were reported to be over 120–150 days (Maguire et al. 1985, 2000). Dowson et al. (1993) also reported that the half-lives of TBT ranged from 360 to 775 days in surface sediments. Furthermore, it is reported that a number of paint chips containing TBT exists in sediment and TBT in paint chips degrade more slowly than that is adsorbed by sediment particles (Stang et al. 1992). It is concluded that no change of TBT concentrations from 1990 to 2000 is due to the stability of TBT in sediments. TPT compounds have been still detected in sediment near small shipyard from the Port of Osaka during 1990–2000 at the value near detection limit.

In the case of plankton and mussel samples, a remarkable decrease of TBT was observed during 1989–1991 and then TBT was decreased moderately (Fig. 3). It is considered that the reduction of TBT levels in plankton and mussels is due to the reduction in water. It is reported that TBT concentrations in water reflect in mussel within 2 or 3 months (Short and Sharp 1989). TBT in adipose tissue of fish decreased moderately between 1991 and 1999 (Fig. 3). The decreasing rate of TBT in fish was lower than those in plankton and mussel samples. TPT concentrations in plankton and mussel have decreased and have not been detected in 2000, while, those in adipose tissues of fish have been still detected.

OT concentrations in Otsuchi Bay have been measured since 1995 (Fig. 4) (Harino et al. 2003). No changes of TBT in water and mussel samples were observed, while, the concentrations in sediment decreased slightly. It is well known that the concentrations of OTs in sediment vary widely (e.g. Harino et al. 1999), because paint chips deposit sometimes in



**Figure 3.** The temporal trend of BTs in sediment, plankton, mussels and adipose tissues of fish from the Port of Osaka. (Harino et al. 2004).

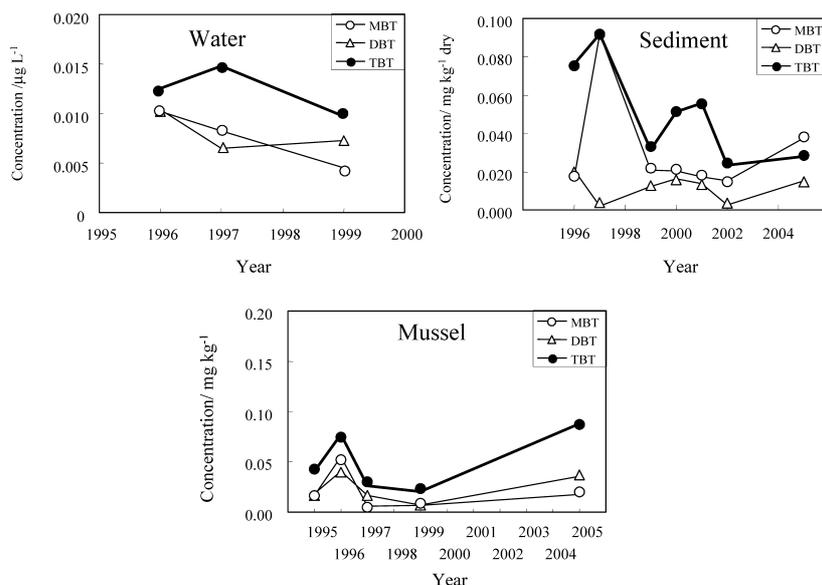


Figure 4. The temporal trend of BTs in water, sediment and mussels from the Otsuchi Bay (Harino et al. 2003).

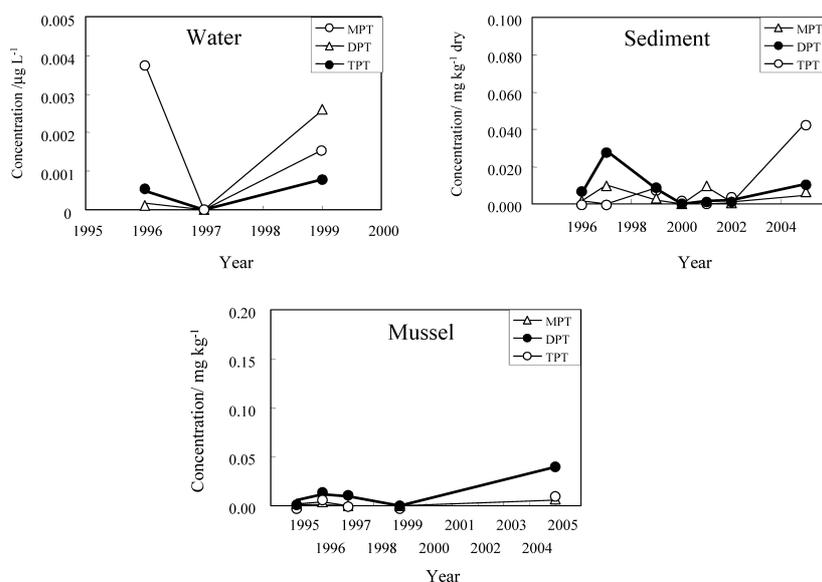


Figure 5. The temporal trend of PTs in water, sediment and mussels from the Otsuchi Bay (Harino et al. 2003).

sediment. The decrease of TBT concentration in sediment may be due to the decrease of the paint chips which deposited to sediment. Continuous survey is needed to clarify the temporal trend of OTs in sediment from Otsuchi Bay. Among BT species, TBT was dominant species in water, sediment and mussels, suggesting a great deal of TBT input to water. TPT was detected at the values near detection limit during the study periods (Fig. 5).

As described above, TBT concentrations in water and biological samples from coastal waters of Japan decreased in about 1990 that was designated as Class 2 Specified Chemical Substances and after that, TBT concentrations were constant. On the other hand, no change of TBT in sediment is observed, in spite of regulation for OTs in Japan. TPT con-

centrations in all compartments of aquatic environment have decreased in this study area of Japan after regulation.

#### Horizontal distribution

The concentrations of TBT and TPT which were reported recently are shown in Table 1 (Inoue et al. 2002, Harino et al. 2003, Sakai et al. 2003, Kobayashi et al. 2007, Ohji et al. 2007) and the characterizations of these coastal areas are shown in Table 2. OT concentrations in water of 7 coastal areas (Otsuchi Bay, Maizuru Bay, The Port of Osaka, Tanabe Bay, Kasado Bay, Hakata Bay and Ariake Bay) of Japan were compared. In spite of the differences of the utilization of these coastal areas, the mean concentrations of TBT and TPT in each coastal area were  $0.011 \mu\text{g l}^{-1}$  and

**Table 1.** TBT and TPT concentrations in water samples from various coastal waters in Japan/water  $\mu\text{g l}^{-1}$ , sediment  $\text{mg kg}^{-1}$  dry, mussel  $\text{mg kg}^{-1}$ 

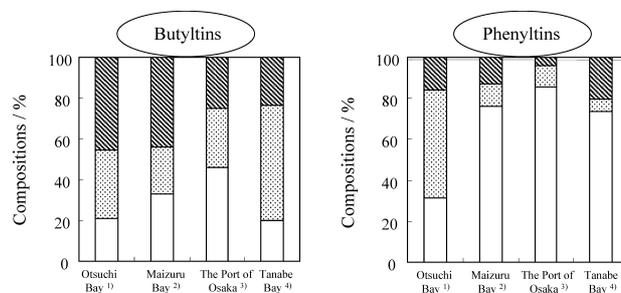
1) Harino et al., 2003, 2) Ohji et al. 2007, 3) Harino et al. 2004, 4) Kabayashi et al. 2007, 5) Inoue et al. 2002, 6) Baasansuren et al. 2004, 7) Sakai et al. 2003

Sample	Area	Year	TBT	TPT
Water	Otsuchi Bay <sup>1)</sup>	1999	0.002–0.022 (0.010)	<0.001–0.001 (0.001)
	Maizuru Bay <sup>2)</sup>	2003	0.004–0.027 (0.011)	<0.0002–0.006 (<0.0002)
	The port of Osaka <sup>3)</sup>	2005	<0.001–0.083 (0.013)	<0.001–0.004 (0.001)
	Tanabe Bay <sup>4)</sup>	2005	0.002–0.014 (0.004)	0.001–0.005 (0.003)
	Kasado Bay <sup>5)</sup>	1998	0.002–0.005 (0.004)	
	Hakata Bay <sup>5)</sup>	1998	0.005–0.038 (0.020)	
	Ariake Sea <sup>5)</sup>	1998	0.003–0.037 (0.018)	
Sediment	Yamada Bay <sup>1)</sup>	2000	<0.001–0.036 (0.009)	<0.001–<0.001 (<0.001)
	Otsuchi Bay <sup>1)</sup>	2005	<0.001–0.201 (0.028)	<0.001–0.088 (0.011)
	Ofunato Bay <sup>1)</sup>	2001	0.006–0.651 (0.124)	<0.001 (<0.001)
	Maizuru Bay <sup>2)</sup>	2003	0.001–0.019 (0.004)	<0.00004–0.005 (0.001)
	Tokyo Bay <sup>6)</sup>	2000	0.002–0.029	<0.0004–0.003
	The port of Osaka <sup>3)</sup>	2005	0.003–0.240 (0.104)	<0.001–0.270 (0.051)
	Kasado Bay <sup>5)</sup>	1998	0.011–0.074 (0.032)	
	Hakata Bay <sup>5)</sup>	1998	0.016–0.450 (0.183)	
	Ariake Sea <sup>5)</sup>	1998	0.007–0.032 (0.014)	
	Suo Nada <sup>5)</sup>	1998	0.003–0.039 (0.025)	
	Uwa Sea <sup>7)</sup>	1999	0.001–0.381 (0.061)	
Mussel	Yamada Bay <sup>1)</sup>	1997	0.012–0.214 (0.103)	<0.001–0.099 (0.025)
	Otsuchi Bay <sup>1)</sup>	2005	0.003–0.381 (0.088)	<0.001–0.096 (0.040)
	Maizuru Bay <sup>2)</sup>	2003	0.001–0.011 (0.004)	<0.00006–0.001 (<0.00006)
	Tanabe Bay <sup>4)</sup>	2005	0.010–0.065 (0.029)	0.079–0.281 (0.168)
	The port of Osaka <sup>3)</sup>	2005	0.011–0.058 (0.037)	<0.001–0.023 (0.010)
	Tanabe Bay <sup>4)</sup>	2005	0.004–0.276 (0.143)	0.005–3.4 (1.7)

**Table 2.** The characterization of coastal area in Japan

Area	Characterization of coastal area
Yamada Bay	Fishing port, Aqua culture
Otsuchi Bay	Fishing port, Aqua culture
Ofunato Bay	Fishing port, Aqua culture
Maizuru Bay	Many timber yards, Navy base.
Tokyo Bay	Industry area. International trading port
The port of Osaka	Industry area. International trading port
Tanabe Bay	Fishing port, Aqua culture
Kasado Bay	Aqua culture, Industrial area
Hakata Bay	International trading port, Commercial area
Ariake Sea	Aqua culture, tidal wetland
Suo Nada	Industry area
Uwa Sea	Aqua culture

0.001  $\mu\text{g l}^{-1}$ , respectively. TBT concentrations were similar in the littoral region around Japan. Compositions of BTs in Otsuchi Bay, Maizuru Bay, the Port of Osaka and Tanabe Bay are shown in Fig. 6. The ratios of TBT in these coastal waters are under 50%, showing that the degradation rate of TBT overcomes the load of TBT to water. On the other hand, Tekeuchi et al. (2004) compared the BTs concentrations in water samples among 18 areas along the Japanese coast. As the result, TBT concentrations in the western Japan were

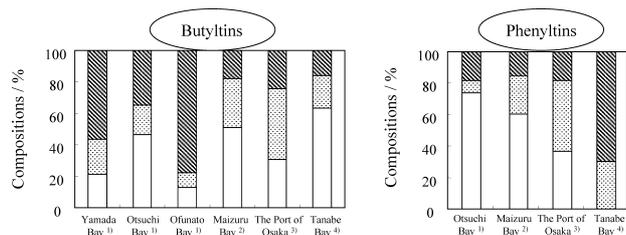
**Figure 6.** Compositions of BTs and PTs in water from adjacent area of Japan

□ Mono, ▨ Di, ▩ Tri

1) Harino et al. 2003, 2) Ohji et al. 2007, 3) Harino et al. 2004, 4) Kobayashi et al. personal communication

higher than those in the Pacific coast of northern Japan, the coast along the Sea of Japan and Tokyo Bay and the adjacent area. TPT concentrations in water from Otsuchi Bay, Maizuru Bay, the Port of Osaka and Tanabe Bay were the values near the detection limit (Table 1) and the ratio of TPT was low among PTs.

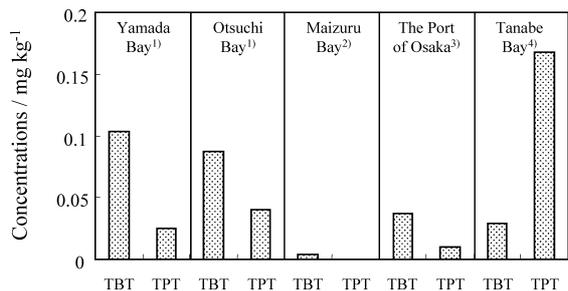
The average of TBT concentration in sediment from Ofunato Bay, the Port of Osaka, Tanabe Bay and Hakata Bay were over 0.1  $\text{mg kg}^{-1}$  dry, suggesting the much more use of this compound in the past year (Table 1), because sediment



**Figure 7.** Compositions of BTs and PTs in sediment from adjacent area of Japan.

□ Mono, ▨ Di, ▩ Tri

1) Harino et al. 2003, 2) Ohji et al. 2007, 3) Harino et al. 2004, 4) Kobayashi et al. personal communication



**Figure 8.** TBT and TPT concentrations in mussel samples from adjacent area of Japan.

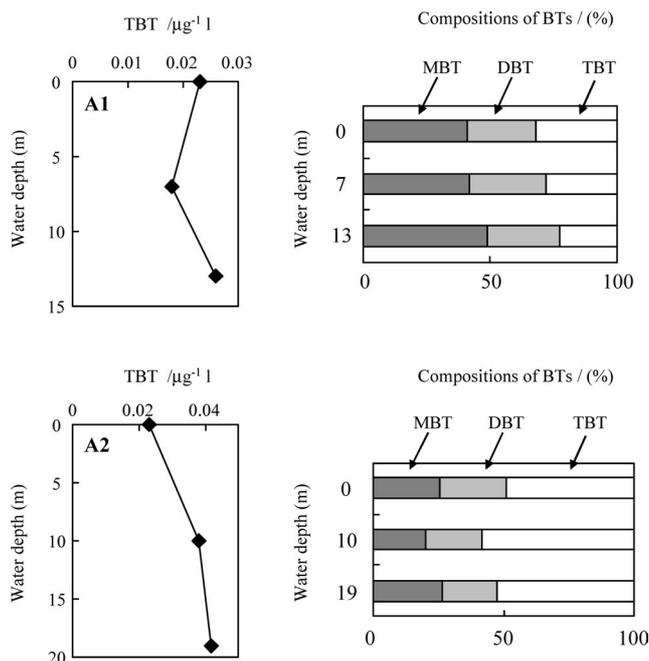
1) Harino et al. 2003, 2) Ohji et al. 2007, 3) Harino et al. 2004, 4) Kobayashi et al. 2007

integrates the load of TBT. Furthermore, among these coastal areas, the highest concentration of TPT in sediment was observed in Tanabe Bay and TPT was dominant compound among PTs (Fig. 7). It is considered that an amount of TPT was also used as an antifouling paint in the past year.

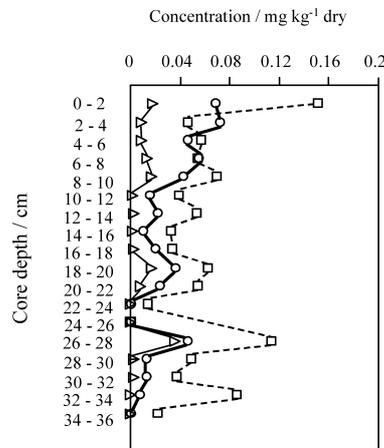
The higher concentrations of TBT in mussel samples from Yamada and Otsuchi Bays at Sanriku coastal area were observed (Fig. 8), suggesting that the load of TBT in these bays has continued, because OTs concentrations in mussel reflects those in water within 2 or 3 months (Short and Sharp 1989). The highest concentrations of TPT in mussel were observed in Tanabe Bay. TPT was detected in all water samples in Tanabe Bay and the higher TPT concentrations in sediment were observed (Fig. 8), suggesting the possibility which TPT has still used as an antifouling paint.

**Vertical distribution**

Harino et al. (1998b) surveyed the BTs concentrations in the surface, medium and bottom layers of water columns from Osaka Bay (Fig. 9). The concentration of TBT and the composition of BTs at stations A1 (water depth 14 m) and A2 (water depth 20 m) were almost uniform in water column. Salinity in surface (A1: 28.36‰, A2: 32.34‰), medium (A1: 30.89‰, A2: 32.34‰) was similar to that in bottom (A1: 31.98‰, A2: 32.34‰) layers, which shows the vertical mixture in water column. Harino et al. (1998b) presume that the similar concentrations of TBT between upper and bottom



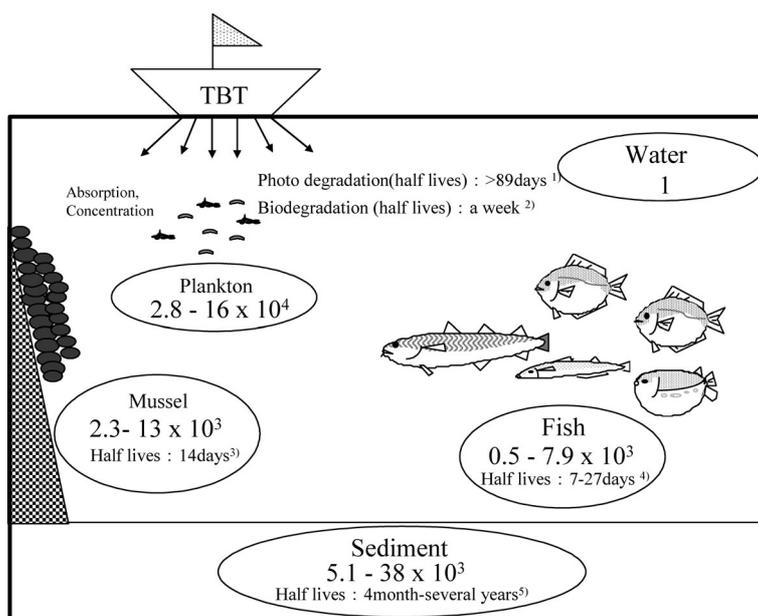
**Figure 9.** Vertical distribution of TBT concentrations and the composition of BTs in water columns (Harino et al. 1998b). A1; The area near the Port of Osaka (water depth 14 m), A2; Central of Osaka Bay (water depth 20 m)



**Figure 10.** Vertical distribution of BTs in sediment core (Harino et al. 1998b). Sampling site: Osaka Bay, ○: TBT, △: DBT □: MBT

layer are due to the vertical mixture in water column.

Figure 10 shows the concentration of BTs in sediment core from Osaka Bay (Harino et al. 1998b). TBT concentration decreased during 0 cm–10 cm in core depths and after that TBT concentration decreased to the values near the detection limit. It is reported that TBT persists for several years within sediment (Maguire et al. 1985, 2000, Dowson et al. 1993). Accordingly, it is possible to estimate the times when release of TBT began in this area from the profile of TBT in sediment core. Namely, the concentration of TBT in the top 10 cm was higher than in the other sediment core and the



**Figure 11.** Behaviour of TBT in aquatic environment.

1) Maguire et al. 1983, 2) Harino et al. 1998b, 3) Laughlin Jr. et al. 1986, Zuolian and Jensen 1989, Page et al. 1995, 4) Yamada and Takayanagi 1992, 5) Dowson et al. 1993, Maguire et al. 1985, Maguire 2000

deposition rate of sediment at this station was calculated to be 3.1 mm/year by  $\gamma$ -ray spectrometer. Consequently, it is estimated that TBT has been roughly used since the 1960. The time estimated from the profile of TBT in sediment core, agreed with the general assertion that TBT has been in use since the 1960's.

### Behaviour of TBT in aquatic environment

Behaviour of TBT in aquatic environment is summarized in Fig. 11. TBT released from ship hull was absorbed to suspended solid or was soluble in water. TBT which was soluble in water, are degraded by sun light and bacteria. Although the half lives of biodegradation are about a week (Harino et al. 1998c), TBT is not degraded by sun light (half lives; >89 days) (Maguire et al. 1983). Furthermore, TBT which absorbed to suspended solid is not degraded. TBT which was soluble in water or was absorbed to suspended solid was accumulated in biota and deposited in sediment. The cross-media partitionings of TBT in Osaka Bay and Otsuchi Bay were calculated using the data of Harino et al. (2003, 2004) by the following formula.

$$K_{sw}, K_{pw}, K_{mw} \text{ or } K_{fw} = Sc, Pc, Mc, Pc \text{ and } Fc/Wc$$

$K_{sw}$ ,  $K_{pw}$ ,  $K_{mw}$  and  $K_{fw}$  are the cross-media partitioning of the sediment–water, plankton–water, mussel–water and fish–water, respectively.  $Wc$ ,  $Sc$ ,  $Pc$ ,  $Mc$  and  $Fc$  are the TBT concentration in water ( $\mu\text{g l}^{-1}$ ), sediment ( $\mu\text{g kg}^{-1}$  dry), plankton ( $\mu\text{g kg}^{-1}$  dry) and mussels ( $\mu\text{g kg}^{-1}$  wet) and fish ( $\mu\text{g kg}^{-1}$  wet).  $K_{sw}$ ,  $K_{pw}$ ,  $K_{mw}$  and  $K_{fw}$  of TBT were ranged of  $5.1 \times 10^3 - 3.8 \times 10^4$ ,  $2.8 \times 10^4 - 1.6 \times 10^5$ ,  $2.3 \times 10^3 - 1.3 \times 10^4$  and  $5.0 \times 10^2 - 7.9 \times 10^3$ , respectively. These results mean that TBT is accumulated easily in biota and sediment. TBT con-

centration in biological samples decreased with reducing of the input of TBT, because the half-lives of TBT in mussel and fish were within a month (Laughlin Jr. et al. 1986, Page et al. 1995, Zuolian and Jensen 1989, Yamada and Takayanagi. 1992). However TBT in sediment will be persisted for a long time (Dowson et al. 1993, Maguire et al. 1985, Maguire 2000). TBT contaminations in sediment will be therefore serious problem in near future.

### An issue in the future

TBT concentrations in water and biological samples reduced in comparison to those in 1990 when the regulation was enacted in Japan. However drastic changes of TBT were not observed in sediment. Generally, TPTs have been decreased after regulation and were not detected in aquatic environment from the most area of Japan. On the other hand, the higher concentration of TPT has still been detected in local site such as Tanabe Bay. Thus, the current status of OTs contaminant varied in aquatic environment of Japan. It is therefore important to monitor the OTs compounds in all sites of Japanese coastal water. Furthermore, TBT persistence in sediment for a long time, may become source of TBT input into water in future. It therefore needs to remove the sediment which was contaminated heavily by OTs. It is likely the environmental issue of OTs contamination will continue after global ban.

### References

- Alzieu, C., Saujuan, J., Deltriel, J. P., Borel, M. 1986. Tin contamination in Arcachon Bay effects of oyster shell anomalies, Mar. Pollut. Bull. 17: 494–498.

- Baasansuren, J., Okazaki, M. and Ando, T. 2004. Butyltin and phenyltin compounds in river/bay sediments of Tokyo. *Soil. Sci. Plant. Nutr.* 50: 159–163.
- Chau, Y. K., Maguire, R. J., Brown, M., Yang, F., Batchelor, S. P. and Thompson J. A. J. 1997. Occurrence of butyltin compounds in mussels in Canada. *Appl. Organomet. Chem.* 11: 903–912.
- Dowson, P. H., Bubb, J., M., Williams, T., P. and Lester, J., N. 1993. Degradation of tributyltin in sediment in freshwater and estuarine marine sediments. *Wat. Sci. Technol.* 28: 133–137.
- Gibbs, P. E. and Bryan, G. W. 1986. Reproductive failure in populations of the dog-whelk *Nucella lapillus*, caused by imposex induced by tributyltin from antifouling paints. *J. Mar. Biol. Assoc. UK* 66: 767–777.
- Harino, H., Fukushima, M., Yamamoto, Y., Kawai, S. and Miyazaki, N. 1998a. Organotin compounds in water, sediment, and biological samples from the Port of Osaka, Japan. *Arch. Environ. Contam. Toxicol.* 35: 558–564.
- Harino, H., Fukushima, M., Kawai, S. and Megumi, K. 1998b. Measurement of butyltin contamination of water and sediment in Osaka Bay, Japan. *Appl. Organomet. Chem.* 12: 819–825.
- Harino, H., Fukushima, M., Kurokawa, Y. and Kawai, S. 1998c. Susceptibility of bacterial populations to organotin compounds and microbial degradation of organotin compounds in environmental water. *Environ. Pollut.* 98: 157–162.
- Harino, H., Fukushima, M. and Kawai, S. 1999. Temporal trends in organotin compounds in the aquatic environment of the Port of Osaka, Japan. *Environ. Pollut.* 105: 1–7.
- Harino, H., Yamamoto, Y., Kawai, S. and Miyazaki, N. 2003. Butyltin and phenyltin residues in water, sediment and biological samples collected from Otsuchi Bay, Japan. *Otsuchi Mar. Sci.* 28: 84–90.
- Harino, H., Yamamoto, Y., Kawai, S. and Miyazaki, N. 2004. Distribution of organotin compounds in aquatic environment—The Port of Osaka, Japan—. In *Proceedings of international symposium on antifouling paint and marine environment* (In SAfE). Shibata, K., Senda, T., (ed.) 61–69. Tokyo.
- Harino, H., Ohji, M., Wattayakorn, G., Arai, T., Rungsupha, S. and Miyazaki, N. 2006. Occurrence of antifouling biocides in sediment and green mussels from Thailand. *Arch. Environ. Contam. Toxicol.* 51: 400–407.
- Inoue, S., Oshima, U., Imada, N. and Honjyo, T. 2002. Tributyltin pollution in coastal areas around northern Kyushu. *Jpn. J. Environ. Toxicol.* 5: 43–50.
- Laughlin, Jr. R. B., French, W. and Guard, H. E. 1986. Accumulation of bis(tributyltin)oxide by the marine mussel *Mytilus edulis*. *Environ. Sci. Technol.* 20: 884–890.
- Maguire, R. J., Carey, J. H. and Hale, E. J. 1983. Degradation of tri-n-butyltin species in water. *J. Agric. Food Chem.* 31: 1060–1065.
- Maguire, R. J. and Tkacz, R. J. 1985. Degradation of the tri-n-butyltin species in water and sediment from Tronto harbour. *J. Agric. Food Chem.* 33: 947–953.
- Maguire, R. J. 2000. Review of the persistence, bioaccumulation and toxicity of tributyltin in aquatic environments in relation to Canada's toxic substances management policy. *Water Qual. Res. J. Canada*, 35: 633–679.
- Midorikawa, S., Arai, T., Harino, H., Ohji, M., Duc, C. N. and Miyazaki, N. 2004. Concentrations of organotin compounds in sediment and clams collected from coastal areas in Vietnam. *Environ. Pollut.* 131: 401–408.
- Ministry of Environment, 2004, *Chemicals in the Environment*. Tokyo, Japan: Environmental Health Department, Environment Agency (In Japanese).
- Ohji, M., Arai, T., Midorikawa, S., Harino, H., Masuda, R. and Miyazaki, N. 2007. Distribution and fate of organotin compounds in Japanese coastal waters. *Water Air Soil Pollut.* 178: 255–265.
- Page, D. S., Dassanayake, T. M. and Gilfillan, E. S. 1995. Tissue distribution and depuration of tributyltin for field exposed *Mytilus edulis*. *Mar. Environ. Res.* 40: 409–421.
- Sakai, H., Kasai, R., Takahashi, S. and Tanabe, S. 2003. Contamination by butyltin compounds in sediments, cultured fishes (*Seriola quinqueradiata* and *Pagrus major*) and pearl oysters (*Pinctada martensii*) collected from Uwa Sea, Japan. *Nippon Suisan Gakkaishi* 69: 10–22 (in Japanese).
- Short, J. W. and Sharp, J. L. 1989. Tributyltin in bay mussel (*Mytilus edulis*) of the Pacific Coast of the United States. *Environ. Sci. Technol.* 23: 740–743.
- Stang, P. M., Lee, R. F. and Seligman, P. F. 1992. Evidence for rapid, nonbiological degradation of tributyltin compounds in autoclaved and heat-treated fine-grained sediment. *Environ. Sci. Technol.* 26: 1382–1387.
- Sudaryanto, A., Takahashi, S., Monirith, I., Ismal, A., Muchtar, M., Zheng, J., Recharadson, B. J., Subramanian, A., Prudente, M., Hue, N. D., Tanabe, S. 2002. Asia-Pacific mussel watch: monitoring of butyltin contamination in coastal waters of Asian developing countries. *Environ. Toxicol. Chem.* 21: 2119–2130.
- Takeuchi, I., Takahashi, S., Tanabe, S. and Miyazaki, N. 2004. Butyltin concentrations along the Japanese coast from 1997 to 1999 monitored by *Caprella* spp. (Crustacea: Amphipoda). *Mar. Environ. Res.* 57: 397–414.
- Yamada, H. and Takayanagi, K. 1992. Bioconcentration and elimination of bis(tributyltin)oxide (TBTO) and triphenyltin chloride (TPTC) in several marine fish species. *Wat. Res.* 12: 1589–1595.
- Zuolian, C. and Jensen, A. 1989. Accumulation of organic and inorganic tin in blue mussel, *Mytilus edulis*, under natural conditions. *Mar. Pollut. Bull.* 20: 281–286.