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MONITORING OF BUTYLTIN COMPOUNDS IN THE AQUATIC ENVIRONMENTS OF THE PHILIPPINES

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ABSTRACT

This article provides a comprehensive review of the contamination by Butyltin compounds (BTs) and its possible implications on the marine and coastal environments of the Philippines. Butyltin compounds were detected in mussels from all the sampling locations investigated as part of the monitoring surveys under the Asia-Pacific Mussel Watch (APMW) Program conducted in the late 1990s. BTs were detected in green mussels collected from all the aquaculture sites in the country, suggestive of considerable widespread pollution by BTs in the coastal waters. Among BTs, TBT was detected at all locations at relatively higher concentrations, whereas the concentrations of DBT and MBT were lower. This could be indicative that green mussels have limited ability of metabolize TBT to DBT and MBT. In addition, this result may be suggestive of fresh TBT inputs into the aquatic environments and the presence of recent sources along the coastal waters. Concentrations of ÓBT in green mussels were reported up to 790 ng/g wet weight in a site where intensive maritime activities are occurring. Mussels from rural areas contained TBT at lower proportions indicating that TBT usage as antifouling agents is minimal. Tolerable average residue level (TARL) for seafood in the Philippines was estimated at 173 ng/g wet weight for an average person weighing 60kg. Concentrations of TBT or the sum of TBT and DBT in some green mussels analysed revealed that some values exceeded TARL level, which could suggest that humans consuming this seafood from areas with high BTs contamination could be at risk from elevated exposure to BTs. Similarly, BTs were detected in all the skipjack tuna collected, suggestive of the widespread contamination even in offshore waters and open seas on a global scale. Skipjack tuna collected from the offshore waters of the Philippines revealed considerable levels (up to 220 ng/g wet weight), with high percentages of BTs in total tin (ÓSn: inorganic tin + organic tin) in the liver tissues. This finding seems to suggest that the anthropogenic BTs represent the major source of Sn accumulation in skipjack tuna. On the other hand, relatively low concentrations of BTs were found in the liver of cetaceans from the coastal waters of the Philippines, which ranged 42-98 ng/g wet weight. Significantly lower hepatic BT concentrations in cetaceans in tropical waters were noted compared with those inhabiting temperate waters proximal to developed nations. This result could imply smaller usage of BTs in the Philippines at least at the present.

Keywords: Butyltin, Green mussel, TBT

INTRODUCTION

Over the last two decades, rapid increase of chemical trade in developing countries in Asia was recognized, which implied greater production and usage of toxic chemicals such as heavy metals, organochlorines and butyltins. These activities could lead to increase exposure of humans and wildlife to these chemicals (Tanabe, 1999). Consequently, environmental problems associated with these persistent organic pollutants became a serious concern, particularly in the aquatic environment.

Since the 1960s, BTs have been widely used for maritime structures, aquaculture activities, lumber preservatives and slimicides in cooling system, and as an effective antifouling agent in paints (Sudaryanto *et al.,* 2002). Its derivatives - dibutytin (DBT) and monobutyltin (MBT) were mostly used as stabilizers in polyvinyl chloride and as catalysts in the production of polyurethane foams, silicones, and in other industrial processes (Fent, 1996). Widespread usage of butyltins motivated the conduct of numerous studies in order to elucidate environmental contamination and impacts. Considerable studies reported adverse

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effects of BTs on non-target marine organisms, such as shell malformation in oysters (Alzieu and Heral, 1984; Beaumont and Budd, 1984; Bryan and Gibbs, 1991). In view of these investigations, aquatic pollution by BTs has been given much attention in many countries, which eventually led to its restriction, particularly the TBT-based antifouling paints, in most developed nations since the 1980s. However, in most developing Asian countries, including the Philippines, such control is yet to be enforced. In addition, an increasing demand for antifouling paints is expected in the Asia-Pacific region (Layman, 1995). Thus, ecotoxicological studies in this region have started giving considerable attention to the aquatic pollution and toxic biological effects by BTs in this region.

Since 1994, monitoring studies in connection with the Asia-Pacific Mussel Watch Program (APMW), aimed at monitoring the pollution in Asian coastal waters have been conducted. Results presented here form part of the collaborative research project among scientists from ASEAN countries, spearheaded by the APMW Project Leader: Prof. Shinsuke Tanabe of the Center for Marine Environmental Studies (CMES) at Ehime University, Japan. Furthermore, in an effort to extend the conduct of continuous and comprehensive monitoring survey of BTs in the ocean environment, widely distributed and globally migrating organisms such as marine mammals and skipjack tuna were also considered for further investigations by the same group of collaborating scientists from the Asia-Pacific region. As one of the participating scientists in these cooperative research investigations, this paper is presented to elucidate the contamination by BTs in the aquatic environments of the Philippines. Moreover, to determine the degree of contamination in these waters, concentrations of butyltin compounds detected were compared with reported values from other regions.

MATERIALS AND METHODS

Samples

Whole soft tissues of green mussels *Perna viridis* from various coastal sites; liver tissues of marine mammals including Fraser's dolphin *Lagenodelphis hosei* and spinner dolphin *Stenella longirostis* from the northeastern part of Sulu Sea; and liver tissues of skipjack tuna collected from the marine waters off the Philippines were considered for the analysis of BTs residues (Fig. 1). Mussel samples were stored in polyethylene bags, kept in a cooler box with dry ice and then

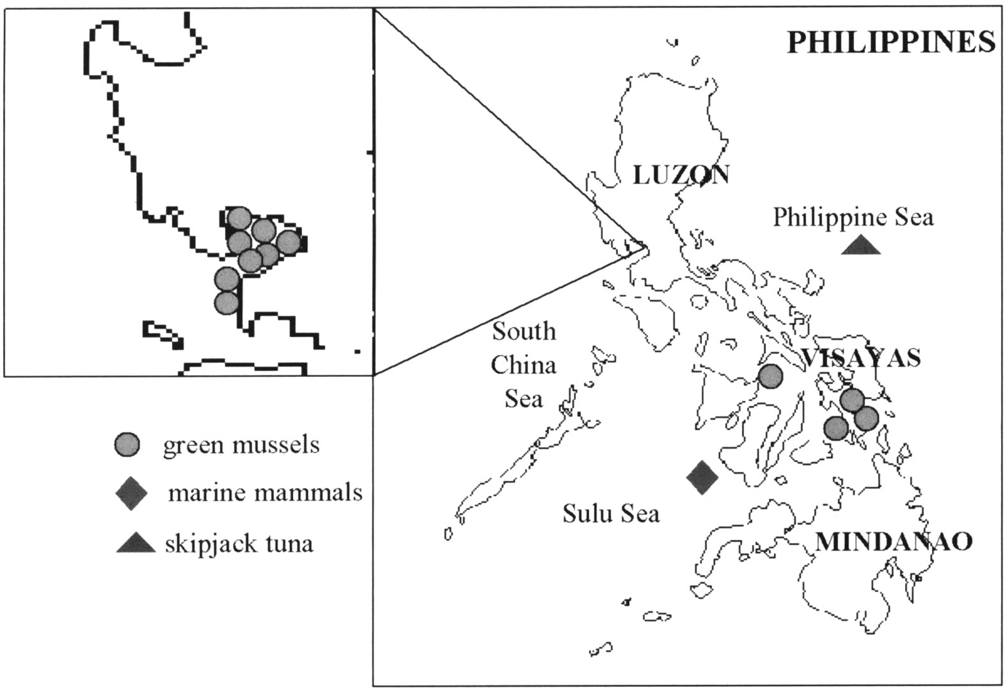

Figure 1. Sites considered for the collection of green mussels, marine mammals and skipjack tuna from the coastal and offshore waters of the Philippines.

immediately kept in a deep-freezer. The frozen mussels were subsequently transported to the laboratory, biometric data measured (Table 1), thawed, scraped clean and shucked. The whole soft tissues were pooled, homogenized, then kept in clean glass bottles. All samples were frozen at -20°C until chemical analysis. On the other hand, after dissection of the marine mammals and skipjack tuna employed for this study, the liver tissues were likewise stored in polyethylene bags at -20°C until analyses.

Chemical analysis

For the whole soft tissues of the green mussels: the analytical method of BTs was conducted based on the procedure previously described (Kan-atireklap *et al.*, 1997). Briefly, the method consists of extraction, propylation, purification and quantification by Hewlett-Packard 5890 Series II gas chromatograph equipped with a flame photometric detector (GC-FPD). Recoveries of BTs from the spiked mussel tissues were 87±14%, 137±20% and 107±25% for MBT, DBT and TBT (n=7), respectively. The detection limit was 3 ng g^{-1} for MBT and 1 ng g^{-1} for DBT and TBT.

For the liver tissues of the marine mammals and skipjack tuna: the chemical analysis of BTs was conducted following the method previously described (Iwata *et al.*, 1995). Concentrations of BTs are reported as nanograms of cation per gram on a wet weight basis, unless otherwise specified. In addition, concentrations were also calculated on a dry-weight basis based on the moisture contents of the samples. In the case of the skipjack tuna, the analytical procedure for total tin (ΣSn) was based on the method described elsewhere (Le *et al.*, 1999) with a slight modification. Detection limit was 10 ng Sn/g on a dry weight basis. The recovery of Sn in this method was checked by analysing a certified material (NIES No. 11), and

Table 1. Biometric data of green mussels collected from coastal waters of the Philippines.

Location	Sampling date	n	Shell length (mm)	Soft tissue weight (g wet wt)
Paranaque, Metro Manila	941122	32	40 (35-50)	1 (0.8-2)
Ermita, Manila Bay	941125	19	47 (40-57)	3 (2-6)
Malate, Manila Bay	941127	32	54 (48-63)	3 (1-4)
Bocaue, Bulacan	941128	41	65 (51-80)	4 (2-8)
Bacoor, Cavite	941130	39	63 (54-80)	4 (2-6)
Bocaue, Bulacan	970411	29	66 (57-80)	8 (5-13)
Bacoor, Cavite	970409	30	77 (63-109)	12 (7-26)
Jiabong, Samar	970404	30	77 (48-100)	7 (2-11)
Villareal, Samar	970405	30	76 (63-86)	8 (6-11)
San Pedro Bay, Leyte	970406	30	78 (62-114)	5 (2-14)
Sapian Bay, Capiz	970407	18	76 (69-87)	5 (4-7)
Samal, Bataan	970408	51	46 (35-53)	3 (2-5)
Malabon, Metro Manila	970410	30	68 (58-80)	10 (5-16)
Pamarawan, Bulacan	980326	15	81 (74-93)	8 (6-10)
Obando, Bulacan	980317	82	58 (17-89)	4 (2-10)
Malabon, Metro Manila	980316	71	63 (41-87)	6 (2-14)
Bacoor, Cavite	980327	43	69 (60-77)	7 (5-19)
San Pedro Bay, Leyte	980321	15	104 (89-117)	17 (13-27)
Jiabong, Samar	980322	46	67 (50-95)	5 (2-13)
Villareal, Samar	980323	51	67 (51-84)	4 (2-8)
Sapian Bay, Capiz	980408	87	64 (41-87)	4 (2-10)

our result (2.5 ± 0.1 μg Sn/g, n=4) agreed well with the certified value ((2.4 ± 0.1 μg Sn/g).

RESULTS AND DISCUSSION

Green mussels. Among the BTs, TBT was detected in most of the sampling sites at relatively high concentrations, whereas the concentrations of DBT and MBT were lower (Table 2). Concentrations of BTs in green mussels collected in 1994-98 ranged from <2 to 51 ng g^{-1} for MBT, <1 to 100 ng g^{-1} for DBT and <1 to 640 ng g^{-1} for TBT. Relatively high TBT concentrations were observed in samples collected from Manila Bay, where maritime activities are quite intensive, considering that is a major shipping traffic area with large harbours. On the other hand, BTs levels were found to be low in green mussels collected from aquaculture areas (ranging from <1 ng g^{-1} to 4 ng g^{-1}), implying minimal usage of BTs for aquaculture activities in the Philippines. Thus, it is probable that the major BTs pollution sources in the country are far sea or commercial vessels that could be using TBT-coated antifouling agents on the ship hulls. These findings also indicate that BTs contamination seem to be widespread along Philippines' coastal waters (Prudente *et al.*, 1999). TBT levels (ranging <1 to 640 ng g^{-1}) found in mussel samples from Philippine waters were comparable with those in Thailand, but were higher than those in other Asian countries such as Malaysia and Hongkong (Table 3). Similar to Thailand (Kan-atireklap et al., 1997), pollution of BTs in the Philippines may be considered to be "high" among Asian developing countries so far reported.

The compositional ratio of butyltin derivatives in *P. viridis* analysed was observed to be in the order of TBT > DBT > MBT. Among BTs, TBT was the dominant compound occupying about 70%

Table 2. Concentrations (ng g^{-1} wet weight) of butyltin compounds (BTs) in green mussels collected from Philippine coastal waters.

Location	Area description	MBT	DBT	TBT	BTs*
1994					
Paranaque, Metro Manila	Urban area, aquaculture area	15	13	76	104
Ermita, Manila Bay	Harbor, shipping line, urban area	47	100	640	787
Malate, Manila Bay	Harbor, shipping line, urban area	51	43	200	294
Bocaue, Bulacan	Agriculture, aquaculture area	5	4	13	22
Bacoor, Cavite	Urban area, aquaculture area	9	8	34	51
Bocaue, Bulacan	Agriculture, aquaculture area	<3	12	34	46
1997					
Bacoor, Cavite	Urban area, aquaculture area	<3	16	43	59
Jiabong, Samar	Agriculture, aquaculture area	<3	3	1	4
Villareal, Samar	Agriculture, aquaculture area	<3	2	28	30
San Pedro Bay, Leyte	Agriculture, aquaculture area	<3	<1	<1	<1
Sapian Bay, Capiz	Agriculture, aquaculture area	<3	1	<1	1
Samal, Bataan	Agriculture, aquaculture area	<3	<1	<1	<1
Malabon, Metro Manila	Urban area, aquaculture area	3	15	44	62
1998					
Pamarawan, Bulacan	Agriculture, aquaculture area	<2.0	2.1	1.7	3.8
Obando, Bulacan	Agriculture, aquaculture area	7.7	5.8	3.9	17
Malabon, Metro Manila	Urban area, aquaculture area	8.1	19	47	74
Bacoor, Cavite	Urban area, aquaculture area	15	18	31	64
San Pedro Bay, Leyte	Agriculture, aquaculture area	4.7	2.8	9.8	17
Jiabong, Samar	Agriculture, aquaculture area	<2.0	<1.3	0.8	0.8
Villareal, Samar	Agriculture, aquaculture area	<2.0	<1.3	0.8	0.8
Sapian Bay, Capiz	Agriculture, Aquaculture area	2.1	1.6	3	6.7
*BTs=MBT+DBT+TBT					

Table 3. Range concentrations (ng g^{-1} wet wt) of MBT, DBT and TBT residues in bivalve molluscs collected worldwide and reported in the literature.

Species	Location (year surveyed)	MBT	DBT	TBT	Reference
Mytilus edulis	East Coast, USA (1989-1990)	ND-28	2-116	2-240	Short and Sharp, 1989
	West Coast, USA (1989-1990)	ND-60	2-148	2-276	Short and Sharp, 1989
	Tokyo Bay, Japan (1989)	20-120	40-450	20-240	Higashiyama *et al.* , 1991
	British Columbia, Canada (1995)	2.4-15	2.7-31	25-153	Stewart and Thompson, 1994
	Coastal Harbor, Canada (1995)	ND-210	ND-416	10-585	Chau *et al.* , 1997
	South and East Coasts, Korea (1997-1999)	9.3-300	19-1100	17-1200	Hong *et al.* , 2002
Mytilus galloprovincialis	West Coast, Portugal (1986)	3.2-169	ND-82	ND-114	Quevauviller *et al.* , 1989
	Western Mediterranean (1996)	10-204	4-1094	1-1151	Morcillo and Porte, 1998
Perna viridis	Thailand (1994-1995)	<3-45	<2-80	3-680	Kan-atireklap *et al.,* 1997
	India (1994-1995)	<3-250	<1-110	<1-150	Kan-atireklap *et al.,* 1998
	India (1998)	<2.2-66	<0.86-150	0.83-570	Sudaryanto *et al.* , 2002
	Indonesia (1998)	1.5-13	<0.58-14	2.2-38	Sudaryanto *et al.* , 2002
	Malaysia (1998)	<2.6-74	<1.0-160	3.5-730	Sudaryanto *et al.* , 2002
	Cambodia (1998)	<2-25	<0.98-37	2.4-88	Sudaryanto *et al.* , 2002
	Vietnam (1998)	<2.1-3.3	<0.86-19	2.1-64	Sudaryanto *et al.* , 2002
	Hongkong (1999)	4.2-90	4.9-76	16-330	Sudaryanto *et al.* , 2002
	Philippines (1994-1997)	<3-51	<1-100	<1-640	Prudente *et al* ., 1999
	Philippines (1997-1998)	<2.0-15	<1.3-19	0.8-47	Sudaryanto *et al.* , 2002

in most locations, while DBT and MBT comprised less than 30 %. These findings conform with the observations made by Kan-atireklap *et al.* (1997) on the limited ability of *P. viridis* to metabolize TBT to DBT and MBT similar to other molluscs. Additionally, continuous input of TBT in the coastal waters of the Philippines may also provide a plausible explanation for the higher ratio of TBT found in mussel samples analysed. It is interesting to note however, that mussels from rural areas contained TBT at lower concentrations and proportions, indicating that TBT usage as antifouling agents in these rural areas seems to be minimal.

Since mussels are one of the commonly consumed seafood items in the country, it is worthwhile to assess possible risk to human health. In an earlier study conducted by Sudaryanto *et al.* (2002), it was pointed out that to assess risk to human health from the consumption of seafood products, tolerable average residue levels (TARLs) as suggested by Belfroid *et al.* (2000) could be calculated. Using the data of average seafood consumption reported by Food and

Table 4. Concentrations (ng g^{-1}wet weight) of butyltins in the liver of cetaceans from Sulu Sea, the Philippine

Species	WBL* (cm)	N M	N F	MBT	DBT	TBT	BTs
Lagenodelphis hosei	221-225	1	1	**15**	**53**	**26**	**94**
Fraser's dolphin				(<4.0-29)	(38-68)	(21-31)	(89-98)
Stenella longirostris	182-192	1	1	**2**	**32**	**21**	**55**
Spinner dolphin				(<4.0-3.1)	(23-41)	(19-23)	(42-67)

*WBL: Whole body length (cm); N:no.of samples.
BTs=MBT+DBT+TBT
Bold:mean concentration. Ranges are given in parentheses.

Agriculture Organization (FAO) of the United Nations, TARL for seafood in the Philippines was estimated at 173 ng g^{-1} wet weight for an average person weighing 60 kg (Sudaryanto *et al.*, 2002). Concentrations of TBT or the sum of TBT and DBT (TBT + DBT) in some of the mussels analysed were found to have exceeded this value, which could be suggestive that people consuming seafood from areas with high butyltin contamination may be at risk from elevated exposure to BTs. Moreover, Sudaryanto *et al.* (2002) evaluated the extent of anthropogenic input of tin by determining the residues of total tin (ΣSn: organic Sn + inorganic Sn) against the BTs residues found in green mussels and reported that the ratio of BTs in ΣSn were higher in the coastal areas where high boating activities were occurring. These observations indicate that anthropogenic sources contribute considerably in tin accumulation in green mussels and that most of the total tin in mussels exists in organic form such as BTs, which further imply that tin compounds originated mostly from anthropogenic sources.

Marine mammals. Relatively low concentrations of BTs were measured in the liver of cetaceans from the Sulu Sea, which ranged at 42-98 ng g^{-1}, wet wt. (Table 4). These findings were similar to the low BTs residues (ranging <1-30 ng g^{-1} wet wt.) found in green mussels from the waters in the Visayas region, which is proximal to the site where these marine mammals were collected. BTs residue levels in these cetaceans compare favorably with those found in cetaceans collected from China (350 to 1200 ng g^{-1}, wet wt), another developing country in Asia (Tanabe *et al.*, 1998). Further, it has been reported that cetaceans inhabiting waters adjacent to developing countries in the tropics and subtropics including the Philippines and India contain significantly lower hepatic BT concentrations compared with those inhabiting temperate waters proximal to developed nations such as Japan (Tanabe *et al.*, 1998). This could be indicative of significant and continuous inputs of BTs in the coastal waters of these developed countries, while smaller usage in developing countries, such as the Philippines, is implied at least at present.

Among the BTs, DBT was predominant in most of the liver samples of these marine mammals, followed by TBT and MBT. This pattern is similar to BTs composition observed in other cetaceans previously studied, which includes harbour porpoises from the Black Sea (Madhusree *et al.*, 1997), bottle-nosed dolphins along the coasts of Italy (Kannan *et al.*, 1996), and stranded cetaceans along the U.S. Atlantic and Gulf coasts (Kannan *et al.*, 1997). These findings are indicative of similar metabolic processes among cetaceans. Interestingly, higher accumulations of DBT/BTs than TBT/BTs were detected in spinner and dolphins collected from the the Sulu Sea, Philippines. In contrast, the same species of spinner dolphins collected from Bay of Bengal, India revealed higher ratio of TBT/BTs

Table 5. Mean and range of butyltins and total tin concentrations green mussels and skipjack tunas collected from Philippine waters.

Species	Location	MBT*	DBT*	TBT*	BTs*	TBT / BTs (%)	SBTs**	SSn**	SBTs / SSn (%)
green mussels	urban coastal waters	**7.9**	**11**	**21**	**40**	**45**	**89**	**130**	**63**
		(<2.0-15)	(2.1-19)	(1.7-47)	(3.8-74)	(23-64)	(9.6-150)	(28-180)	(28-107)
green mussels	rural coastal waters	**2.2**	**1.7**	**3.6**	**6.3**	**76**	**17**	**47**	**26**
		(<2.0-4.7)	(<1.3-2.8)	().8-3)	(0.8-17)	(45-100)	(1.6-74)	(17-74)	(4.2-64)
skipjack tuna	offshore waters	**1.9**	**35**	**150**	**180**	**80**	**220**	**230**	**88**
		(< 1.8-5.7)	(16-66)	(62-200)	(78-270)	(75-86)	(95-310)	140-320	(68-100)

Bold:mean concentration; ranges are given in parentheses.

*ng/g wet wt

**ng Sn/g dry wt

than DBT/BTs (Tanabe *et al.*, 1998). The reason for the apparent difference in BTs composition among the spinner dolphins collected from India and the Philippines remains unclear. However, higher maritime activities in the Bay of Bengal than in Sulu Sea could be a plausible explanation.

Skipjack Tuna. Mean and range concentrations of BTs and ΣSn in the liver of skipjack tunas collected from offshore waters of the Philippines and the green mussels from various coastal waters is shown in Table 5. Concentrations of BTs in the liver tissues of skipjack tuna collected off Philippine waters in December 1997 ranged at 78-270 ng/g wet weight. Among BTs, TBT was detected at relatively higher concentrations, whereas DBT and MBT were lower. This pattern conforms to those observed in green mussels, which indicate continuous inputs of TBT into the aquatic environment and the presence of recent sources in the offshore waters.

Similar to the pattern observed in green mussels, high percentage of BTs in total tin (ΣSn: inorganic tin + organic tin) were found in the liver tissues of the tunas, suggestive that anthropogenic BTs represent the major source of Sn accumulation in skipjack tuna. As reported by Ueno *et al.* (2004), BTs occupied higher percentages in ΣSn concentrations in skipjack tuna collected from offshore waters around Asian developing countries having high concentrations of BTs, which suggested that hepatic Sn exists primarily in organic form as BTs that originate from anthropogenic sources.

CONCLUSION

Significant butyltin concentrations in green mussels collected from the coastal areas of the Philippines revealed contamination along nearshore marine waters. Relatively high BTs concentrations were observed in green mussels collected from areas with intensive maritime activities, suggestive that antifouling paints could be the major TBT source. While low TBT levels found in green mussels from aquaculture areas imply minimal usage of BTs for aquaculture activities. Similarly BTs residues found in skipjack tuna collected from offshore waters of the Philippines indicated BTs contamination and presence of recent butyltin pollution sources. High percentages of BTs in ΣSn found in the liver of the skipjack tuna suggest anthropogenic BTs as major source of tin accumulation in these animals. While the considerable BTs levels found in the liver tissues of marine mammals collected from Sulu Sea, Philippines imply that BTs accumulate at measurable levels in the liver of higher trophic marine animals. Considering the unregulated usage of TBT and the possible increasing demand for TBT-containing paints in the Philippines, contamination by BTs in its aquatic environments may become serious in the future. It is therefore necessary to conduct continuous monitoring in order to make appropriate decisions on the future issues of butyltin contamination in the aquatic environments.

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