

# Contamination status of butyltin compounds in Indonesian coastal waters

Inneke RUMENGAN<sup>1\*</sup>, Madoka OHJI<sup>2</sup>, Takaomi ARAI<sup>3</sup>, Hiroya HARINO<sup>4</sup>, Zainal ARIFIN<sup>5</sup> and Nobuyuki MIYAZAKI<sup>6</sup>

<sup>1</sup> Faculty of Fisheries and Marine Science, Sam Ratulangi University, Manado, Indonesia  
\*E-mail: innekerumengan@hotmail.com

<sup>2</sup> Institute of Symbiotic Science and Technology, Tokyo University of Agriculture and Technology, Fuchu, Tokyo 183–8509, Japan

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

<sup>4</sup> Osaka City Institute of Public Health and Environmental Sciences, 8–34 Tojo, Tennoji, Osaka 543–0026, Japan

<sup>5</sup> Research Center for Oceanography—Indonesian Institute of Science, Jl Pasir Putih I, Jakarta, Indonesia

<sup>6</sup> Center for International Cooperation, Ocean Research Institute, The University of Tokyo, 1–15–1 Minamidai, Nakano, Tokyo 164–8639, Japan

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**Abstract**—Indonesia as an archipelago country relies on shipping for multipurpose transportation, and therefore has a potential risk of butyltin (BT) based biocide contamination. The available data were limited to BT accumulation in surface sea water, sediment, bivalves and fish collected sporadically at restricted areas in 1998 and 2001–2005 showing the presence of ‘hot spots’ in the country. BT concentrations in sediments (dry wt), bivalves (wet wt) and fish (wet wt) in hot spots (big harbors in Bitung, Jakarta, Surabaya and Medan) were in range of 209–425, 23–320, 5–64 and 0.41–19 ng g<sup>-1</sup>, respectively, mostly in order of TBT > DBT > MBT. Occurrence of imposex in gastropods (*Thais* spp) has been also detected in 1989 in a remote island, Ambon Island. Among locations surveyed, commercial harbors and marina are highly contaminated, and in particular BTs levels in sediments of harbors are hundreds folds that in the fisheries areas, indicating the major input of BTs are from antifouling paints of ships. Sediments are of great concern as the deposited BTs could be potential sources in the future, even if after the global ban had been effectively implemented. Moderate levels of BTs in aquaculture areas have revealed the usage of BTs as biocides in aquaculture industries as well. This review shows the BT contamination has been in critical levels, since no restriction applied in this country, fresh input of BTs into the marine environment perhaps continues, and could expand and threaten continuously aquatic ecosystem in Asia–Pacific region, considering the geographical position of Indonesian waters. Monitoring program and ecotoxicological risk assessment with more range of ecosystem compartments are obviously necessary.

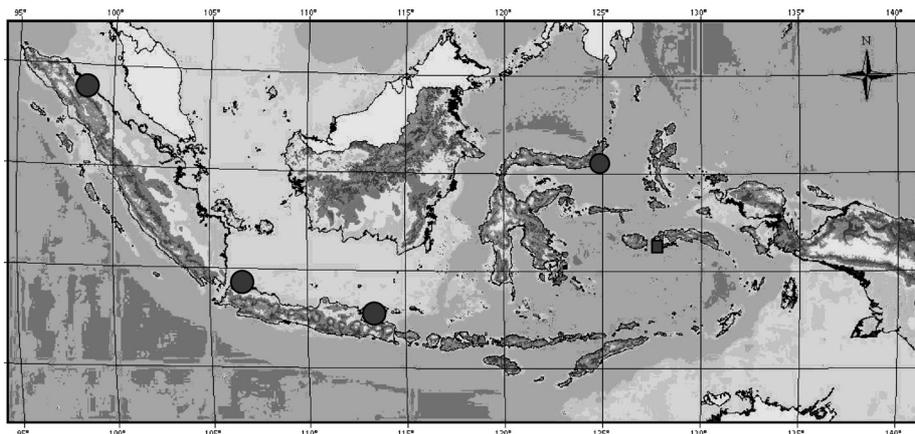
**Key words:** Indonesia, organotin compounds, imposex, aquatic ecosystem, critical level

## Introduction

Indonesia is the largest archipelago in the world, situated at the confluence of the Pacific and the Indian Oceans, and bridging two continents, Asia and Australia. It consists of more than 17,508 islands, about 6,000 of which are inhabited, with a population of with over 234 million people (ADB, 2004, Soegiarto and Stel 1998). Indonesia shares borders with Singapore, Malaysia, and the Philippines to the north and Australia to the south across narrow straits of water (Fig. 1). Indonesian waters surrounded by more than 81,000 km-long coastline, holding nearly 37% of all known marine species. The Indonesian seas form the onliest tropical inter-ocean link, connecting the reservoir of the warm surface water mass of the western Pacific with the eastern Indian Ocean, while transforming it through vertical mixing

and air-sea interaction on its way (Soegiarto and Stel 1998). As the other ASEAN countries, coastal waters of Indonesia are among the most productive and biologically diverse in the world, encompass some of the world’s richest areas in terms of both biodiversity and potential economic activity (Pomeroy et al. 2007).

The Indonesian marine environment constitutes an area of about 400,000 km<sup>2</sup> with a composite structure of environmental conditions, local areas of upwelling, strong wind-driven and tidal currents, and nutrient inputs from rivers or human activities (Tomascik et al. 1997). The marine ecosystem is influenced by a monsoonal system, with a pattern of surface currents varying during the southeast and northwest monsoon. As Indonesian waters form a bridge between two oceans and two continents, it implies ecologically vulnerable coastal and marine ecosystems. Coastal ecosystems are among the most biologically productive, but in the other



**Fig. 1.** Map of Indonesia showing the hot spots of butyltin contamination in 1998, 2005 (●), and imposex occurrence in 1989 (■).

hand they are the receiver of the contaminants washed off the land, and could be sources of contaminants to the adjacent seas, especially those connected to Indian Ocean, because it has been identified since Snellius Expeditions in 1980s that transport within Indonesian waters is directed into the Indian Ocean because there is continuous flow into the ocean with a magnitude of 1.0 to 2.5 Sv (1 Sv (Sverdrup) is equivalent to  $10^6 \text{ m}^3 \text{ s}^{-1}$ ) (Pariwono et al. 2005).

As Eisler (1989) reviewed that 36 of the 260 known organotin compounds are potentially very hazardous to natural resources, especially butyltins compounds (BTs) to aquatic biota. BTs is the most well known organotin compounds, including the mono-, di- and tributyltins (MBT, DBT, TBT). The most concern BTs is TBT, not only used as a marine biocide in anti-fouling paint, but also in wood treatment and preservation, antifungal action in textiles and industrial water system (Extoxnet 1996). There has been a great deal of public concern focused on the toxicological and ecotoxicological aspects of TBT compounds. TBT are capable of causing adverse biological effects at levels far below that any previously reported marine pollutant. Hundreds papers worldwide had linked the compounds to adverse environmental or health effects. The most worrisome are 'profound reproductive effects' coupled with diminished marine-species populations. They persists in the environment, bioaccumulate through the food web, and pose a risk of causing adverse effects to human health and the environment. Under aerobic conditions, TBT take place 1 to 3 months to degrade, but in anaerobic soils, this compound will persist for more than 2 years (Extoxnet 1996).

Indonesia as an archipelagic nation, relies on shipping as an important mode of transportation of natural resources, goods and people, with about 538 ports of which 131 are open for international trade. More than a decade ago, there were already 344 inter-island vessels with 843,000 DWT (Dead Weight Ton), 1119 local transportation vessels (180,000 DWT), 3974 vessels (209,000 DWT) for transporting people, and 27 international vessels (347,000 DWT) in

Indonesia waters (Soegiarto and Stel 1998). Therefore, it is a great concern of potential risk of organotin-based paints in ships. Large quantities of TBT and other organotins still enter the sea directly from the anti-fouling paints on ships' hulls, especially without restriction and ban applied. Unlike in many countries, a number of national and regional bans on the use of this chemical have been applied since 1980s. Efforts are even now under way through the United Nation body, the International Maritime Organization, Marine Environment Protection Committee and other international bodies, to institute a total TBT ban worldwide. A treaty for this global ban is ratified within days, probably January 2008. Vessels with active TBT on the hull will be refuse entry into ports or face large fines.

Widespread contamination of BTs along the coastal waters of Asian developing countries including Indonesia in 1990s has been reported several authors (Ellis and Pattisina 1990, Evans et al. 1995, Sudaryanto et al. 2002, 2004, 2005, Midorikawa et al. 2004b) using the data gained from samples collected between 1989–2003, for some ecosystem compartments such as sediment, bivalves, gastropod and fish. This paper attempts to review the status of organotin contamination in coastal waters of Indonesia, in particular for butyltin compounds, because no data on the other species of organotin compounds available. The use and regulations, and urgent requirements for further monitoring programs are discussed in order to evaluate the global ban of using TBT and other organotin biocides from the year 2008. This review may not contain all available ecotoxicological data on organotin application in the region, but it is expected to promote and foster the dissemination of information on the ecotoxicological effects of organotin compounds.

### Uses and regulation

Indonesia is one of the major tin-producing countries (WHO 2005), even as the world's largest producer of tin in 2003 (33%) (ATSDR 2005). Tin has been used as a structural material and as a chemical for a wide variety of purposes. In-

organic tin compounds in form of Tin(IV)chloride ( $\text{SnCl}_4$ ) is often used as the starting material for production of organotin compounds. Interestingly, the consumption of inorganic tin compounds is lower than that of organotin compounds. (ATSDR 2005, WHO 2005). Organotin compounds have many applications, which include use as stabilizers in PVC (polyvinyl chloride) catalysts in chemical reactions, glass coatings, agricultural pesticides, biocide in marine antifouling paints and wood treatment and preservatives (Batt 2006). The most concerned organotin compound is triorganotin such as tributyltin and triphenyltin compounds, which are used as industrial biocides, agricultural chemicals, wood preservatives, and marine antifouling agents.

BTs have been used within aquaculture industry in Indonesia, as molluscicides, which considered by GESAMP (1997) should be curtailed. This compounds which commercially named Brestan<sup>®</sup>, Aquatin<sup>®</sup> and Thiodan<sup>®</sup> are used to eliminate mollusks prior to stocking of shrimp ponds (GESAMP 1997), but no published data on this topic specifically related to pond aquaculture. It is possible that acute toxic residues will remain in pond sediments for months or longer after treatments. Evaluation of the environmental and human health risks have not been reported. Not only Indonesia, but probably other Asian countries face the same problem, as reported by Sudaryanto et al. (2002) that relatively high concentrations of BTs were found in mussels from aquaculture areas in Hong Kong, Malaysia and Thailand. They predicted that an increase in TBT usage occur in aquaculture, as the economic improvement in those areas.

Indonesia with the 81,000-km coastal line are very potential for development of fisheries and coastal aquaculture, as important source of employment, providing jobs to an estimated six million people engaged in capture fisheries and aquaculture (ADB 2004). Therefore, the country should have regulatory controls and/or documentation of the chemicals used by the industry. Unfortunately, there is no information on the quantities of any organotin compounds actually applied, while the demand for antifouling paints has been estimated still increasing in the Asia-Pacific since 1998 (Jamari 1999).

Antifouling paints containing biocides including tributyltin are classed as biocidal products and are regulated like pesticides in many countries. As one of the signatories of Stockholm Convention, an international legally binding agreement on persistent organic pollutants, which is entered into force, on 17 May 2004 the Indonesian Government has issued some regulations to restrict and manage the hazardous waste, such as Government Regulation No. 19/1994, No. 12/1995, and further No. 74/2001. In the last regulation, only Aldrin, Chlordane, DDT, Dieldrin, Endrin, Heptachlor, Mirex, toxaphene, Hexachloro-benzene and PCBs are prohibited to be utilized. However, organotin-based pesticides are not specifically listed. Syafrul (2003) listed at least 25

regulations related to hazardous chemicals have been issued by 2002 in the country, but no one directly related to hazardous organotin restriction. The only regulation refer to tributyltin is of the Health Ministerial Decree No. 907/2002 concerning the requirements and controls of drinking water quality, in which the maximum concentration of TBTO allowed is  $10 \mu\text{g l}^{-1}$ . Dissemination on the biocidal butyltin compounds, and their persistence in the environment and capacity to build up in the aquatic food chain are needed. Those are the main key issues for restriction of those hazardous compounds.

Although no data on the amount of the consumption of these compounds is available, but considering its archipelagic form, which is situated between the two continents and the two oceans, international and domestic shipping is obviously significant threat to the marine ecosystem, though this may be more significant around Java, where the majority of the population reside. From the world's 30,000 commercial ships used the poison tributyltin (Greve 2007), it can imagine how many Indonesian ships and/or other international ships pass the Indonesian waters.

Throughout the 1980s many countries worldwide began restricting the use of TBT paints. The Organotin Antifouling Paint Control Act was passed in the US, which restricted the use of TBT paints to vessels greater than 25 meters in length (Batt 2006). Major U.S. and European makers voluntary stopped producing TBT in January 2001, and its presence in marine organisms is declining, but it continues to be widely used in much of Asia including Indonesia. Presently, the International Maritime Organization (IMO) has proposed a worldwide total ban for TBT presence on vessel hulls as of January 1, 2008. A year after the TBT treaty is ratified, neither the ships of ratifying countries nor foreign vessels that enter their waters will be allowed to have TBT on their hulls unless a sealant has rendered it inert. Ships found in violation will be put on an international blacklist and barred by other ratifying countries. It will invoke the TBT ban and blacklist system betted by hefty fines, in January 2008 (Greve 2007)

### **Accumulation of butyltin compounds in marine ecosystem**

#### *Water*

In general, it is very few data concentrations of butyltin compounds in seawater of Indonesian waters. The available data is only from northwest of coastal Java (Razak 2004, 2005), and Gangga Island and Bitung Harbor in North Sulawesi waters (our study). For reference, the nearest country, Peninsular Malaysia, the concentration of TBT in seawater ranged from 3.4 to  $20 \text{ ng l}^{-1}$  in unexposed areas (i.e. away from boat and shipping activity), and from 30 to  $281.8 \text{ ng l}^{-1}$  in coastal areas with high boat and ship activities (Tong et al. 1996). Concentration of BTs in hot spot could be hundreds fold that in unexposed area. The levels in the unexposed areas are comparable to the concentrations of TBT in

seawater collected in industrial areas nearby Maizuru Bay, Japan, which ranging from 3.9 to 27 ng l<sup>-1</sup> (Ohji et al. 2006a). TBT is not persistent in most natural waters because it is susceptible to natural degradation processes and it absorb onto particulate matter (Hashimoto et al. 1998). U.S. EPA (2003) has set the new saltwater chronic criterion for TBT, the criterion to protect saltwater aquatic life from chronic toxic effects is 7.4 ng l<sup>-1</sup>, while that acute toxic effects is 420 ng l<sup>-1</sup>. Based on data of a decade ago, contamination status of TBT in seawater in Malaysian waters and most likely Indonesian waters was already in chronic toxic levels. No more investigation on the current contamination status, but with no restrictions and ban applied, it could be on the acute toxic levels nowadays.

#### Sediment

Table 1 shows widespread contamination of butyltins in Indonesian coastal waters, especially in Sulawesi, Java and Sumatera, the islands with much more human residents than the other islands. The incredible amount of TBT deposited, up to 190 ng l<sup>-1</sup> dry wt (59% of total BTs), in Belawan Medan, North Sumatera (Sudaryanto et al. 2005), which was the third biggest harbor at that time, suggesting new fresh input of TBT into the harbor, but the trend of data is corresponding to high TBT concentrations in mussel in harbor and port areas (Sudaryanto et al. 2000, 2002). For comparison, the range of BTs in sediments worldwide is 10 to 1,000 ng g<sup>-1</sup> dry wt, while TBT concentrations in other ASEAN Countries were in range of 4 to 4,500, <0.7 to 216.5, and 0.89 to 34 in Thailand, Malaysia and Vietnam, respectively as summarized by Midorikawa et al. 2004a), but could be as high as 12,400 ng g<sup>-1</sup> dry wt in a hot spot area in USA in 1990–1992 (Sudaryanto et al. 2005). Comparatively, the average of TBT concentrations in sediment from selected areas in Japan was over hundreds ng g<sup>-1</sup>, for example in Otsuchi Bay ranging from 10 to 640 ng g<sup>-1</sup> dry wt (Harino et al. 1998), and 2 to 966 ng g<sup>-1</sup> dry wt in harbor area of Osaka city (Harino et al. 1999), and more recently Ohji et al. (2006a) measured the concentrations TBT in sediment collected in Maizuru Bay ranging from 1.2 to 19 ng g<sup>-1</sup> dry wt. Incredible high level of TBT in big harbors and fishing ports clarifying the source of input from ships moored there. TBT was the predominant compound among BT derivatives, which indicates that the TBT was just newly released into the waters, and/or it has been slowly metabolized in those compartments. It is, alternatively, caused by the lower uptake efficiency, bioavailability, or greater excretion rates for DBT and MBT.

TBT is identified as a contaminant of potential concern in the marine sediments. It degrades much more slowly in sediment and is likely to persist in sediments at concentrations which cause adverse biological effects (U.S. EPA 1999), having an estimated half live of 2.5 years (Hashimoto

et al. 1998), and in heavily contaminated sediments it becomes even more persistent. Levels in sediments are commonly 1000 times greater than levels in overlying waters (Allsopp and Johnston 2000). Therefore, U.S. EPA (1999) recommends that when TBT is a contaminant of concern in sediment, interstitial water concentrations should be measured and that sediment toxicity testing or *in situ* or laboratory bioaccumulation testing should be conducted to confirm the ecological significance of measured interstitial water concentrations.

#### Fish

The field data of fish as shown in Table 1 ranging from 3.3 to 150 ng g<sup>-1</sup> wet wt of BTs concentrations with the order of BTs was TBT>DBT>MBT (Ueno et al. 2004; Sudaryanto et al. 2005). This trend shows that newly TBT input in seawater have bioconcentrated to a considerable extent in the muscle tissues of various fish. Among fish, the pelagic fish accumulated BTs higher than demersal species, which is attributable to the direct uptake of BTs released into water column before deposition into the sediments (Sudaryanto et al. 2004). Although no fish samples were analyzed from the other areas of the country, they may contain high concentrations of BTs. Kannan et al. (1995) measured BTs concentrations in tissues of fish collected from Bogor, West Java in November 1991, such as big-eyed scad (*Selar crumenophthalmus*) and deep bodied crucian carp (*Carassius cuvieri*). They reported the presence of BTs derivatives in the order MBT>DBT>TBT. Approximately 80% of the BTs present in the tissue were in the form of MBT, which suggests that TBT degrades in the aquatic environment and in biota (Jamari 1999).

The main concerns regarding the exposure of fish to BTs are focused on their potential for immunotoxicity, hepato toxicity, neurotoxicity, and developmental toxicity (Snoeji et al. 1987). The chronic toxic effects of TBT on the immune system of fish occur at a few hundred ng l<sup>-1</sup> in seawater or even lower concentrations (Kannan et al. 1995). Wester and Canton (1987) determined in their laboratory work that the limit of 'no-observed-effect' concentration of TBTO for guppy was 10 ng l<sup>-1</sup>. Occasionally, however, higher TBT concentrations in the water, up to 11,700 ng l<sup>-1</sup>, may cause hazard to sensitive aquatic organisms when exposed for long periods.

Ohji et al. (2006b) considered, however, that TBT accumulated not only in marine or freshwater fish but also in diadromous fish such as masu salmon, *Oncorhynchus masou* (Brevoort). They demonstrated that the TBT concentrations in sea-run masu salmon were in range of 6.2 to 7.2 ng g<sup>-1</sup> wet wt, significantly higher than those in freshwater-resident ones, but much lower than that in some fish collected from Indonesia (Table 1). The proportion of BTs in each fish might be affected by the presence of BTs in the water envi-

**Table 1.** Accumulation of BTs in some compartments of marine ecosystem in Indonesian waters.

Compartment	Location	Period of sampling	Concentration					Reference
			MBT	DBT	TBT	$\Sigma$ BTs		
Surface water (ng l <sup>-1</sup> )	Gangga Island Bitung Harbor	March 2005	<0.1 <sup>-6</sup>	<0.1	14-23	209-425	This study	
			10	<1	52			
	Bitung Harbor	March 2005	49-76	2-3	158-346	209-425	This study	
			1	2	89			107
	Gangga Island	March 2005	2	2	1	5	This study	
			<1-3.5	3.2-32	19-506			
Jakarta Bay	July 2003	<1-1.2	3-16	26-649	Razak 2005			
	Nov 2003	<0.5-2.2	0.5-2.7	<0.5-12				
Banten Bay	Aug 2003	<0.5-1.0	<0.5-1.1	<0.5-4.2	Razak 2005			
	Oct 2003	11-37	4.6-87	2.3-190				
Sediments (ng g <sup>-1</sup> dry wt)	Big harbors (Jakarta, Surabaya and Medan)	July 1998	8.0-32	13-27	9.6-10	32-69	Sudaryanto et al. 2005	
		July 1998	<2.5	<2.7	0.51-0.82	0.51-0.82	Sudaryanto et al. 2005	
	Fishing port, urban center, industry (Jakarta)	July 1998	<2.5	<2.7	0.63-1.2	0.63-3.2	Sudaryanto et al. 2005	
Aquaculture (Lampung, Panimbang)	July-August 1998	July-October 1998	<2.5	<2.7	0.51-0.82	0.51-0.82	Sudaryanto et al. 2005	
		July-August 1998	<2.5	<2.7	0.63-1.2	0.63-3.2	Sudaryanto et al. 2005	

Table 1. (Continued)

Compartment	Location	Period of sampling	Concentration				Reference
			MBT	DBT	TBT	$\Sigma$ BTs	
	Big harbors (Jakarta, Surabaya and Medan)	July 1998	2.8–13	<0.58–14	2.2–37	5–64	Sudaryanto et al. 2000, 2002
	Fishing port, urban center, industry (Jakarta)	July 1998	7.6–11	6.7–8.6	13–38	27–58	Sudaryanto et al. 2000, 2002
	Fisheries areas (Cirebon, Makassar)	July–August 1998	2.3–2.5	1.1–2	2.5–2.9	6.1–7.2	Sudaryanto et al. 2000, 2002
Mussel, <i>Perna viridis</i> (ng g <sup>-1</sup> in wet wt)	Aquaculture (Jambi, Lampung, Panimbang)	July, August, October 1998	<1.5–2.6	<0.58–3	3.4–4.3	3.7–9.9	Sudaryanto et al. 2000, 2002
Arkshells, <i>Anadara antiquata</i>	Jakarta	March 2003	1.6–2.1	2.9–3.4	14–14	—	Midorikawa et al. 2004
	Jakarta	March 2003	2.7–11	9.5–35	9.7–37	—	Midorikawa et al. 2004
Fish (various sp, muscle tissues) (ng g <sup>-1</sup> in wet wt)	Bogor	November 1991	<5.6–10	0.41–4.8	<0.13–3.7	0.41–19	Kannan et al 1995
Fish (various sp, tissues) (ng g <sup>-1</sup> in wet wt)	Big harbor (Jakarta)	July 1998	2.9–14	3.8–18	9.7–52	21–84	Sudaryanto et al. 2005
	Fishing port (Cirebon)	August 1998	1.2–3.4	<1.5–3.4	2.1–18	3.3–25	Sudaryanto et al. 2005
	Aquaculture (Panimbang)	August 1998	0.9–4.2	1.5–6.2	1.4–12	4.2–18	Sudaryanto et al. 2005
Fish (skipjack, liver) (ng g <sup>-1</sup> in wet wt)	Offshore waters	January 2001	<1.8	4.7–61	7.9–88	13–150	Ueno et al. 2004

ronment. Tropical diadromus fish like eels in Indonesian is potentially used as bioindicator for assessment of hazardous risk of BTs and its bioavailability and biomobility. Diadromous fish has a risk of TBT exposure, their accumulation patterns differing according to their life histories. Furthermore, the reproductive toxicity of TBT might induce population disturbance (Ohji et al. 2006b). Adverse effects of BTs and other pollutants in hot spot areas in Indonesian waters have never been studied, although sudden death of fish population is annually happened in some areas, such as just recently (November 2007) in Bali Island.

### Bivalves

Bivalves such as mussels have been used as bioindicators for monitoring trace toxic substances in coastal waters. Sudaryanto et al. (2002) observed *Perna viridis* in western Indonesian waters, Sumatra and Java, which were in range of 3.7 to 64 ng g<sup>-1</sup> wet. The compositional ratio of BTs was in order TBT>MBT>DBT, which suggests fresh input at that year of 1998, particularly in aquaculture area in Jambi, a fishing port and marina in Jakarta and harbor in Surabaya, in range of 58–100% (Sudaryanto et al. 2002).

Midorikawa et al. (2004b) also measured the TBT concentrations of mussels, *P. viridis* and arkshells *Anadara antiquate*, which was collected in March 2003 from Jakarta, Indonesia, were 14 and 19 ng g<sup>-1</sup> wet wt, respectively. These levels were higher than those from uncontaminated sites in Vietnam and Thailand, but lower than a contaminated site in Vietnam. Composition of BTs compounds were in the order of TBT>DBT>MBT, suggesting fresh input of TBT to the coastal environment in the country. They also measured the TPT levels, in range of not detected to 23.1 ng g<sup>-1</sup> wet wt. Comparetively, Ohji et al. (2006a) measured the concentrations of TBT in blue mussels *Mytilus galloprovincialis* collected in Maizuru Bay, Japan ranged from 0.77 to 11 ng g<sup>-1</sup> wet wt, respectively, lower than those in Indonesia.

### Occurrence of imposex in gastropods

A major concern with TBT is its ability to cause imposex (the superimposition of male anatomical characteristics on females) in a variety of species. Imposex has been observed in 45 species of snails worldwide, with definitive laboratory and field studies implicating TBT as the cause in cosmopolitan species (U.S. EPA 2003). As shown in Table 2, the imposex cases are found in gastropods belonging to the family Muricidae (Mollusca: Prosobranchia) in Ambon, Indonesia and some areas of Singapore, Malaysia and Japan. It is clear that Ambon Island surrounding waters has been contaminated with BTs since a decade ago, based on the relatively high RSP Index (up to 46.7%) of particular gastropods such as *T. luteostoma*. Ambon Bay has been used extensively as harbor for fishing, cargo and passenger boats.

Imposex was recorded in four gastropod species in

Ambon (Ellis and Pattisina 1990, Evans et al. 1995), each of which is apparently site specific, and therefore, none of them found in Singapore, where about 20 species appear to susceptible to TBT and capable of displaying imposex (Tan 1997). Interestingly, *T. luteostoma*, which was previously found in 1989 by Ellis and Pattisina (1990) with severe symptom of imposex, was no longer recorded in 1993 by Evans et al. (1995). Within only four years a part, there has probably been lost due to the highly contaminated with TBT. Females with imposex exhibit abnormal reproduction and/or irregular embryological development and consequently further extinction of population (Tan 1997). It remains for future studies for clarification.

Table 2 also shows that the same species from different area showing different RPSI (relative penis size index), suggesting that gastropod hormonal disturbance implies the degree of contamination status of BTs. However, there is a tendency that different species from same area having different RPS indices indicating the species specific responses of the gastropods toward the BTs contamination level. Widespread contamination in Asian countries in that period of time has been revealed by discovering cases of imposex not only in Singapore and Malaysia (Tan 1997), but also in *T. clavigera* from oyster marine culture areas in Taiwan (Hung et al. 2001).

### Toxicity effects

Very limited laboratory works on toxicity effects of TBT and TPT are accessible. In the study of Rumampuk et al. (2004a), three species of microalga *Tetraselmis tetrahele*, *Nannochloropsis oculata* and *Dunaliella* sp, the important phytoplankton in many hatcheries, were shortly exposed to TBT concentrations of 0.1, 0.5 and 1 µg l<sup>-1</sup>. The chlorophyll a and b contents of *T. tetrahele* in all treatments were higher than those in controls, even be doubled in 1 µg l<sup>-1</sup> of TBT. *N. oculata* and *Dunaliella* had a slightly higher chlorophyll a and b contents in the lowest TBT concentration tested (0.1 µg l<sup>-1</sup>) than those in control, but as the TBT concentration increased their chlorophyll contents decreased. Three levels of TBT tested are within the range of the no observable effect concentration (NOEC) for *T. tetrahele*, while the lowest observable effect concentrations (LOEC) for *N. oculata* and *Dunaliella* are between 0.1 to 0.5 µg l<sup>-1</sup>. Among the three species, *N. oculata* has a highest sensitivity towards TBT.

Toxic effects of triphenyltin (TPT) on the marine alga, *Eucheuma denticulatum* obtained from Nain Island, North Sulawesi, Indonesia have been examined in laboratory condition by Rumampuk et al. (2004b). The algae were exposed to different concentrations of TPT between 0–30 µg l<sup>-1</sup> with 5 µg l<sup>-1</sup> interval. After two-weeks, the aniline blue stained tissue of alga *E. denticulatum* showed morphological changes at all concentrations tested. The medullary cells of

**Table 2.** Occurrence of imposex for gastropods in Ambon Island, Indonesia and other selected areas of Singapore and Japan.

Species	Location	Period	RPSI (%)	Imposex frequency (%)	Reference
<i>T. luteostoma</i>	Ambon Bay	July–August 1988	0.29–46.7	64–93	Ellis and Pattisina 1990
	Ambon City		20.8–26.2	30–100	
	Port Dickson, Malaysia	April 1989	0.001	100	
<i>T. kieneri</i>	Ambon Bay	July–September 1993	0.5–6.3*	41.3–85.1*	Evans et al. 1995
	South-east coast, Ambon Island		<0.01	6.7	
<i>T. savignyi</i>	South-east coast, Ambon island		0.63	8.1–28.7*	
<i>V. turbinellus</i>	South-east coast, Ambon Island		0.76–0.84*	7.7–8.1*	
<i>T. bitubercularis</i>	Pasir Panjang, Singapore	September 1991	22.6	—	Tan 1997
	The mouth of Fort Road monsoon drain Singapore	October 1992	1.6	—	
	The mouth of Fort Road monsoon drain	November 1992	2.5	—	
<i>T. clavigera</i>	Pasir Panjang, Singapore	September 1991	22.6	—	Tan 1997
	The mouth of Fort Road monsoon drain Singapore	October 1992	13.6	—	
	The mouth of Fort Road monsoon drain	November 1992	22.7	—	
	Nicol Drive, Changi, Singapore	February 1993	18.8	—	
	Arasaki and Misaki, Kanagawa Pref. Japan	April 1990	83–88	—	
	Kohama Beach, Shizuoka, Japan	April 1990	37	—	
<i>T. jubilaea</i>	Fort Road monsoon drain, Singapore	October 1992	22.3	—	Tan 1997
	Fort Road monsoon drain, Singapore	November 1992	35.5	—	

\* The range is calculated from mean  $\pm$  standard errors.

alga exposed to the TPT concentrations from 5 to 15  $\mu\text{g l}^{-1}$  reduced in size compared to those in control. Cell walls of some medullary cells damaged at 20  $\mu\text{g l}^{-1}$  and all cell totally broken at 25 and 30  $\mu\text{g l}^{-1}$ . The pattern of zonately dividing tetrasporangium had not occurred anymore in alga exposed to TPT even at the lowest tested concentration (5  $\mu\text{g l}^{-1}$ ). Reproductive cells of alga is more sensitive than somatic cells, indicating that possible population extinction could happen in TPT contaminated area. Further toxicity

tests using TBT is essential.

Scientific articles that imply that BTs may affect human immune system, or hormonal disruptor have been published, but no report from Indonesia. As recently reviewed by Batt (2006), there was a study dealt with the effects of butyltins on natural killer cell activity in human blood samples. BTs are detected in the blood of human volunteers. Sources of butyltin are hypothesized as coming from PVC compounds and fish contaminated with TBT from antifoulant paints.

Therefore, potential toxicity risks of organotin compounds on humans are obviously important to assess. In order to address many of the environmental concerns over organotins, it is urgently required to establish a database of technical and environmental information relating to toxicity effects of organotin compounds for comparison with the field contamination status in the country.

### Challenges: Monitoring Program

Marine organisms have the ability to accumulate contaminants from the environment where they live at much concentrations and, at the same time, showing much less spatial and temporal variability. On the other hand, ecosystems are under the pressure of complex mixtures of contaminants whose affects are not always simple to assess. In fact, it has been argued that a full understanding of ecotoxicological processes must consider an integrated multilevel approach (Picado et al. 2007). Therefore, it is required to develop a long term monitoring program within national level framework. This framework would be a major contribution to the national database for policy makers. In addition, because organotin contamination in biotic and abiotic compartments of coastal ecosystem vary geographically, and as an archipelagic country, the national level programs should be defined according to local specificities, namely the existing hydrodynamic data and ecological relevant condition. The first step should be to investigate the spatial variation of the contaminants in order to define sites for long term monitoring. Several sites among hot spots should be included, particularly in big harbors in five islands, Sumatera, Java, Kalimantan, Sulawesi and Papua. The next step is to conduct temporal distribution, where sampling should be annually for at least three to five years as Picado et al. (2007) propose for the assessment of environmental quality of coastal waters. Ideally contaminants should be analyzed in main ecosystem compartments such as water, sediment, plankton, benthic organisms, pelagic organisms mainly fish including migratory fish such as eel and tuna. It is important to establish a cause and effect relationship between contamination levels and biological effects, and to assess potential transfer through food chain.

Regarding the use of organotin compounds in aquaculture, mechanisms need to be put in place and enforced for the registration and control of aquacultural chemicals to protect the environment and human health and to ensure the sustainable growth of the aquaculture industry. Evaluation of risks associated with organotin molluscicides is complicated by the lack of quantitative data on their use. It is necessary to obtain field data useful for quantifying risk such as the effluent concentrations of the chemicals and the nature and extent of biological response in receiving waters (GESAMP 1997). The need for mechanisms at national level to compile and maintain records on the quantities of the chemicals should be put in priority. As Indonesia not only shares borders with

Southeast Asian countries, but also the impact of any contamination, these countries should come into agreement to increase efforts to reduce pollution and strengthen the conservation of their resources.

### Concluding Remarks

The contamination status of BTs in Indonesian waters has been in the critical level, as shown by the sporadically available data between 1989 and 2003. Although butyltin concentrations in some compartments (sediments, mussel and fish) were lower than those reported for other countries in the period of time the specimen collected, but since no restriction applied in this country, it is predictable that fresh input of butyltins into the marine environment continues during this decade. With the contamination status and probably more severe condition in recent years, the toxic levels could represent a major threat to marine biodiversity, as well as to humankind. The extent contamination in fish has raised questions of the level of exposure of the fish-eating human population in the country. There is obviously an urgent need to draw this to public attention and ensure that the implications are fully understood.

To date no registration procedures for antifouling paints exist under pesticide/biocide laws in the country. As the organotin compounds will be banned in 2008 worldwide, the intention to implement baseline studies should come into actions soon. Even after global ban, the deposited butyltin compounds in sediments persist, therefore monitoring program and ecotoxicological risk assessment with more range of ecosystem compartments including seawater and plankton are obviously necessary.

Indonesian Government needs to implement pollution reduction strategies as quickly as possible by enacting relevant regulations in national level. However, as increase of concerns regarding possible environmental risks and human health effects associated with the term 'organotin compounds', it is inappropriate to categorize all tin compounds as having equivalent toxicological and ecotoxicological profiles. Clear distinction must be drawn between those used as biocides and pesticides, and those as polymer additives which exhibit no biocidal properties, before the government enacting a domestic regulation to implement the global ban.

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