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Concentrations of organotin compounds in aquatic biota in coastal waters of Johor, Malaysia

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Research Article

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Abstract

The concentrations of organotin (OT) compounds, butyltin (BT) and phenyltin (PT), in aquatic organisms from Merambong and Tinggi Island, Malaysia, which differ in industrial and economic activities, were measured. Tributyltin (TBT) compounds among BTs ranged from 2.9–28 and <0.1–21 ng g^{-1} ww in aquatic organisms from Merambong and Tinggi Island, respectively. Triphenyltin (TPT) compounds among PTs ranged from <0.1–25 ng g^{-1} ww and <0.1–61 ng g^{-1} ww in aquatic organisms from Merambong and Tinggi Island, respectively. A survey of OT concentrations among the species of OT compounds revealed that the concentrations of BTs and PTs in crabs and clams were high while those in fish were low. Correlation analysis with stable nitrogen isotopes suggest that OT compounds did not accumulate in aquatic organisms through the food web. The measurement of OT compounds in tissues and organs of fish revealed that BT concentrations in liver were higher than those in muscle and that BTs were detected in eggs of fish.

Introduction

Organotin (OT) compounds have been utilized as antifouling biocides since 1960. Higher concentrations of OT were detected in coastal areas around the world (Chiavarini *et al.*, 2003; Harino *et al.*, 2005*a*, 2005*b*). Those compounds have been of great concern due to their adverse effects on non-target marine organisms, such as oyster shell malformations and imposex (Laughlin & Linden, 1985; Gibbs & Bryan, 1986; Bryan & Gibbs, 1991; Gibbs *et al.*, 1991). In 2008, the International Convention on the Control of Harmful Antifouling Systems on ships (AFS Convention), which prohibited the use of OTs as active ingredients in antifouling systems for ships, was ratified by the International Maritime Organization (IMO). After the ban on the use of tributyltin (TBT) compounds, the concentrations of OT compounds in aquatic environments must be monitored in order to evaluate the effect of the regulation. However, there are few reports on the measurement of organotin compounds concentrations (Lee *et al.*, 2015; Ruiz *et al.*, 2015).

Aquaculture is a main industry in Southeast Asia, and aquatic resources such as farm-raised shrimp are exported. Plantation-produced palm oil is also a major export industry in Southeast Asia. These two main drivers of the economy in Southeast Asia are made possible by the climate and natural environment there. However, the economic development means that trade will flourish, and active trade leads to OT pollution. Before the IMO regulations, there were many papers on the concentrations of OTs in the Association of Southeast Asian Nations (ASEAN). For example, Midorikawa *et al.* (2004) reported on OT concentrations in in Vietnam. The concentrations of TBT in sediment and clams were in the ranges of 0.89–34 ng g^{-1} dry weight (dw) and 1.4–56 ng g^{-1} wet weight (ww), respectively. Harino *et al.* (2006) reported that concentrations of TBT in sediment and green mussels from Thailand ranged from 2–1246 ng g^{-1} dw and 4–45 ng g^{-1} ww, respectively. Sudaryanto *et al.* (2002) reported widespread contamination with butyltin (BT) compounds along the coastal waters of developing Asian countries by the measuring the concentrations in green mussels (*Mytilus edulis*). Among Southeast Asian countries, Malaysia has been undergoing particularly rapid economic growth in the past two decades. Therefore, there are concerns about contamination with organotin compounds in Malaysia. Tong *et al.* (1996) reported that TBT in water samples from Peninsular Malaysia was in the range of <3.4–20 ng l^{-1} and that imposex may be caused in gastropods. Hashimoto *et al.* (1998) reported concentrations of BT compounds along the Strait of Malacca that were high enough to cause deleterious effects on aquatic organisms, such as imposex in gastropods. Sudaryanto *et al.* (2004) reported the concentrations of BT compounds in Malaysia; they detected BTs in all samples, ranging from 3.6–900 ng g^{-1} ww, 3.6–210 ng g^{-1} ww, and 18–1400 ng g^{-1} dw in mussels, fish and sediments,

respectively. Harino *et al.* (2009) detected higher concentrations of TBT and triphenyltin (TPT) compounds in sediment and green mussel (*Perna viridis*) from Peninsular Malaysia; the maximum concentrations of TBT found in green mussel were near the tolerable average residue level (TARL). Thus, higher concentrations of TBT were detected in aquatic environments in Malaysia. Therefore, attention must be paid to OTs contamination in Malaysian coastal waters.

This study elucidates the status of OT compound contamination in aquatic animals in Malaysia after the IMO ban on the use of such compounds. The study also characterizes the bioaccumulation of OT compounds in aquatic organisms by examining the relationships between the OT concentrations and the trophic level, using stable nitrogen isotope $\delta^{15}N$ to evaluate bioaccumulation of OTs, or biological measurement of aquatic organisms. The results form the basis of a discussion about the biological impact of OTs on aquatic biota in coastal waters of Malaysia and provide additional information about contamination in coastal ecosystems.

Materials and methods

Sampling description

Malaysia is located in the centre of Southeast Asia and consists of two geographic regions divided by the South China Sea: Peninsular Malaysia and Malaysian Borneo. Among Southeast Asian countries, Malaysia has become famous as an exporter of electronic parts, and both aquaculture and palm oil plantations have also become major industries there. Many seagrasses and mangrove areas have been destroyed by the large number of factories, farms and trading ports constructed around the southern part of Peninsular Malaysia. Malaysia has also become famous as a resort recently, because many areas face the sea.

We focused on Merambong and Tinggi Island, which have different industrial bases. Merambong is located between the southern part of Peninsular Malaysia and Singapore (Figure 1). Although the area has many factories, a big trading port, and reclamation of the waterfront, there are some rich natural environments with mangrove and seagrass areas. The seagrass beds are ~1.3 km in length. Tinggi Island is located at the east coast of Johor, and the tourism industries are its main economic activities. Most of the coastal area in Tinggi Island, featuring long coastlines with white sandy beaches is used as a resort area.

To confirm the status of organotin compound contamination of aquatic animals in the seagrass area of Malaysia, many aquatic animals were taken as samples from Merambong and Tinggi Island. From Merambong, on 7–8 September 2012, 4 species of clam, 3 of crab, 1 of cuttlefish, 1 of mollusc, 3 of sea cucumber, 2 of sea star, 2 of shrimp, 2 of snail, and 11 species of fish were taken; on 27–29 May 2013, 6 species of fish were taken (Table 1). From Tinggi Island on 7–8 September 2012, 2 species of clam, 2 of crab, 1 of cuttlefish, 4 of sea cucumber, 1 of sea star, 1 of sea urchin, 1 of shrimp, and 15 of fish were taken (Table 2). These animals were dissected immediately after collection. All samples were brought back to Japan in a cold box and stored in a freezer at −20°C until chemical analysis.

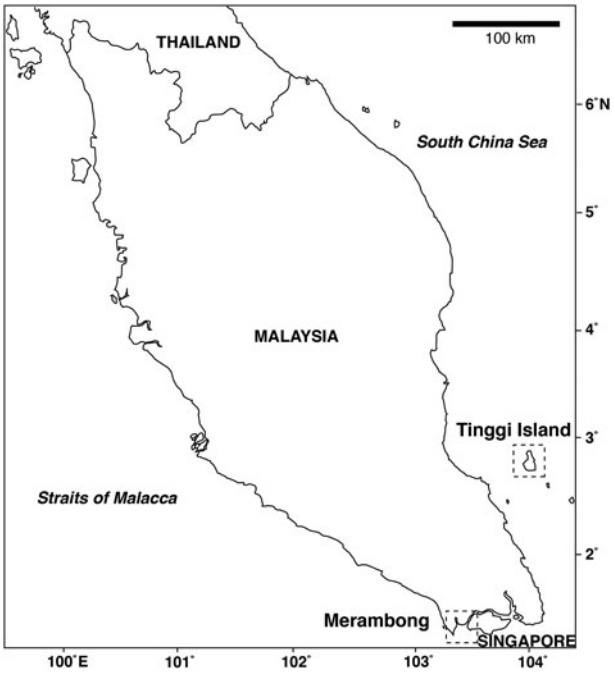

Fig. 1. The sampling sites of biota collected from Merambong and Tinggi Island, Malaysia.

Chemical analysis

The method used to determine the OTs in biological samples was based on that of Harino *et al.* (2012), with some modification. Two grams of sediment sample or plant sample was cut by scissors and put in a centrifuge tube. One hundred micrograms of mixed acetone solution containing 1 μg ml^{-1} each of monobutyltin trichloride (MBTCl)-d_9, dibutyltin dichloride (DBTCl)-d_{18}, tributyltin monochloride (TBTCl)-d_{27}, monophenyltin trichloride (MPTCl)-d_5, diphenyltin dichloride (DPTCl)-d_{10}, and triphenyltin monochloride (TPTCl)-d_{15} was added to the centrifuge tube as a surrogate standard. The mixture was shaken with 10 ml of 1 M HCl-methanol/ethyl acetate (1/1) for 10 min. After centrifugation for 10 min, the residue was again extracted with 10 ml of 1 M HCl-methanol/ethyl acetate (1/1). After centrifugation, the combined supernatants and 30 ml of saturated NaCl solution were then transferred to a separatory funnel. The analytes were extracted twice with 15 ml of ethyl acetate/hexane (3/2) solution, and the organic layer was combined. Fifty millilitres of hexane was mixed into the organic layer, and the mixture was left to stand for 20 min. After removal of the aqueous layer, the organic layer was dried with anhydrous Na_2SO_4 and concentrated up to trace level. The analytes were diluted with 5 ml of ethanol, 5 ml of acetic acid – sodium acetate buffer (pH 5.0), and 10 ml of distilled water, followed by ethylation by shaking with 1 ml of 5% $NaBEt_4$ for 30 min. The solution containing ethylated OTs was saponified with 10 ml of 1 M KOH–ethanol solution by shaking for 1 h. Forty millilitres of distilled water and 10 ml of hexane were added to the solution, and the mixture was shaken for 10 min. The ethylated OT residue in the aqueous layer was extracted again by shaking for 10 min with 10 ml of hexane. The combined organic layers were dried with anhydrous Na_2SO_4. After being concentrated up to 1 ml, the solution was cleaned by a Florisil Sep-Pak column (Waters Association Co. Ltd). The analytes were eluted with 10 ml of 5% diethyl ether/hexane. All eluting solvent was collected in a bottom flask. The solution was concentrated up to 0.5 ml after the addition of tetrabutyltin (TeBT)-d_{36} and tetraphenyltin (TePT)-d_{20} as internal standards. The final solution was concentrated up to 0.5 ml.

A gas chromatograph (6890A series, Agilent Technologies) equipped with a mass spectrometer (5973N, Agilent) was used for analysis of the OTs. The separation was carried out in a capillary column coated with 5% phenyl methyl silicone (J&W Scientific Co., 30 m length × 0.25 mm i.d., 0.25 μm film thickness). The column temperature was held at 60°C for the first 2 min, then

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Table 1. Biological measurement of species of biota in seagrass area of Merambong, Malaysia, in 2012 and 2013

Year	Taxon	Species	TL/SL/CW/ML (cm)	Wet Wt (g)	Year	Taxon	Species	TL/SL/CW (cm)	Wet Wt (g)
2012	Clam	Mactridae sp.	5.8–8.0	31–74 (4)	2012	Fish	*Arius oetik*	9.4–18.2	16–107 (2)
		Pinctada sp.	7.2	54			*Atule mate*	14.2	53
		Pinna sp.	24.3	153			*Lagocephalus lunaris*	14.2	94
		Solen sp.	9.1	27			*Gymnothorax* sp.	22	9
	Crab	*Charybdis* sp.	5.8–5.9	3.2 (2)			*Plotosus canius*	19	69
		Portunus pelagicus	7.3–12.1	25–138 (5)			*Monacanthus chinensis*	7.8–13	15–78 (8)
		Pleocyemata sp.	ND	6–36 (5)			*Scomberoides* sp.	33.5	511
	Cuttlefish	*Sepia recurvirostra*	7.5	60			*Sillago ingenuua*	17.5	75
	Mollusc	Aplysidae sp.	ND	37–41 (2)			Tetrarogidae sp.	7.1	15
	Sea cucumber	*Holothuria fuscopunctata*	ND	419			*Triacanthus biaculeatus*	13.3	85
		Actinopyga lecanora	ND	34–113 (5)			*Tylosurus* sp.	71–75	801–1032 (2)
		Holothuria sp.	ND	9–19 (7)	2013	Fish	*Drepane punctata*	16	249
	Sea star	*Astropecten* sp.	ND	13–23 (3)			*Echeneis naucrates*	36–37	253–288 (2)
		Protoreaster nodosus	ND	375 (2)			*Eleuteronema tetradactylum*	42.1	1428
	Shrimp	*Acetes* sp.	2.8	0.16 (30)			*Leptomelanosoma indicum*	38	1005
		Charybdis feriata	4.5	265 (2)			*Oxymonacanthus longirostris*	13.5–14.3	88.5–94.4 (2)
	Snail	*Melo melo*	9.8	106			*Sillago sihaa*	15.2	57.2
		Strombus canarium	5.4–6.3	11–31 (9)					

TL, total length; SL, shell length; CW, carapace width; ML, mantle length.
Parentheses show the numbers of individuals.

Table 2. Biological measurement of species of biota in seagrass area of Tinggi Island, Malaysia, in 2012

Taxon	Species	TL/SL/CW/ML (cm)	Wet Wt (g)
Clam	*Anadara* sp.	6.7	82
	Pinna sp.	25.5–30.5	77–205 (16)
Crab	*Charybdis granulata*	6.1–7.8	45–94 (4)
	Charybdis orientalis	5.6	26
Cuttlefish	*Sepia* sp.	11.5–12.2	102–104 (2)
Fish	*Abudefduf sexfasciatus*	8.3	23
	Arothron immaculatus	9.4–10.3	50–65 (2)
	Chaetodon octofasciatus	6.4	11
	Choerodon sp.	14.1	83
	Epinephelus bleekeri	8.7	16
	Lethrinus genivittatus	6.6–7.5	8–12 (2)
	Lethrinus lentjan	6.1–18.0	70–152 (4)
	Lutijanus lutijanus	9.8	25
	Luthanus fulviflamma	15.3	120
	Lutjanus vitta	11.6	46
	Pentapodus setosus	8.0	11
	Pomacentrus adelus	8.1	26
	Scolopsis aurata	8.4–13.7	15–69 (5)
	Scolopsis taenioptera	13.4	63
	Valenciennea sexguttata	7.5	6
Sea cucumber	*Bohadschia sp.*	ND	1145
	Cucumaria frondosa	ND	348–485 (2)
	Holothuria edulis	ND	53–190 (5)
	Stichopus vastus	ND	1096
Sea star	*Protoreaster nodosus*	ND	445–584 (2)
Sea urchin	*Diadema sp.*	6.3	60
Shrimp	Alpheidae sp.	1.0	5 (7)

TL, total length; SL, shell length; CW, carapace width; ML, mantle length.
Parentheses show the numbers of individuals.

increased to 130°C at 20°C min^{-1}, 210°C at 10°C min^{-1}, 260°C at 5°C min^{-1} and 300°C at 10°C min^{-1}. Finally, the column temperature was held at 300°C for 2 min. The interface temperature, ion source temperature and ion energy were 280°C, 230°C and 70 eV, respectively. Selected ion monitoring was operated under this program. The monitoring ions of 235 (233) for MBT, 261 (263) for DBT, 263 (261) for TBT, 253 (255) for MPT, 303 (301) for DPT and 351 (349) for TPT were used to quantify the concentrations of OTs. The parentheses show the qualifier ions. One microlitre of the sample was injected by splitless injection. The concentrations of OTs in this study are expressed as Sn^{4+}.

Evaluation of the analytical procedure

The quality of the data obtained by the analytical procedure for OTs and alternative biocides was examined. When 1 µg of OTs were spiked to 1 g of biological sample, the recovery rates of the OTs were in the range of 90–109% and the relative standard deviations (RSDs) were in the range of 8–12%. The detection limits of each OT, based on a signal-to-noise ratio of 3, was 0.1 ng Sn g^{-1} wet wt.

Stable nitrogen isotope ($\delta^{15}N$)

The concentrations of stable nitrogen isotope, $\delta^{15}N$, were measured because $\delta^{15}N$ is used to evaluate bioaccumulation. The method used to determine nitrogen-stable isotopes in plant samples was based on that of Vu *et al.* (2017). The samples were kept frozen (∼ −20°C) for a few days after collection and then were moved to a laboratory and dried in an electric oven at 60°C for 6 h. They were kept dry until analysis at room temperature. The samples were powdered using a pestle and mortar. Subsamples were placed in ultrapure tin capsules, and the samples were burned in an elemental analyser (Flash EA, Thermo Fisher Scientific, Waltham, MA, USA). Combustion gases continuously moved through a flow controller (ConFlo III, Thermo Fisher), and then the stable nitrogen isotope compositions were detected with a mass spectrometer (Delta plusXP, Thermo Fisher). L-alanine was used as the working standard. Stable isotope ratios were expressed in δ notation (parts per thousand, ‰) as deviations from international standards according to the following equation:

$$\delta X = (R_{\text{sample}}/R_{\text{standar}}d{-}1) \times 1000$$

where X represents ^{15}N and R represents isotope ratios $^{15}N/^{14}N$. R_{sample} and R_{standard} are the isotope ratios of the sample and the international standard, atmospheric N_2, respectively.

Statistics

Differences between data were analysed using the Mann−Whitney U-test, and the significance of the correlation coefficient and that of the regression slope were tested by Spearman's rank correlation coefficient (Sokal & Rohlf, 2003).

Results

The concentrations of OTs in aquatic organisms

The concentrations of the total BTs (∑BTs = TBT + DBT + MBT) in aquatic organisms from Merambong and Tinggi Island were found to be in the ranges of 8.2–151 and 3–109 ng g^{-1} ww, respectively (Table 3, Figures 2 and 3). A comparison of the concentrations of ∑BTs between Merambong and Tinggi Island revealed ∑BTs of 32.4 ± 21.4 (Mean ± SD) ng g^{-1} ww and 29.2 ± 13.0 ng g^{-1}, respectively, indicating no significant differences ($P > 0.5$) (Table 3, Figures 2 and 3).

Mean TBT levels were in the range of 5.4–11 ng g^{-1} ww in aquatic organisms from Merambong (Table 3, Figure 2). The concentrations of TBT were compared among species of aquatic organisms, revealing similar levels. The ratio of DBT and MBT, which are degradation products of TBT, were higher than those of TBT in each aquatic organism from Merambong (Figure 4). Mean TBT levels were in the range of <0.1–21 ng g^{-1} ww in aquatic organisms from Tinggi Island (Table 3, Figure 3). The

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Table 3. Concentrations of BTs and PTs (ng g^{-1} wet wt) in biota from Merambong and Tinggi Island, Malaysia, in 2012

Sampling location	Taxon	TBT	DBT	MBT	ΣBTs	TPT	DPT	MPT	ΣPTs
Merambong	Clam	8.3	11	13	33	3.2	5.3	2.3	11
		(5.3–10)	(6.3–17)	(2.5–46)	(14–74)	(<0.1–6.4)	(2.7–7.9)	(<0.1–10)	(2.7–24)
	Crab	11	15	12	37	5.0	6.5	3.6	15
		(5.2–28)	(6.2–39)	(1.2–38)	(13–105)	(<0.1–20)	(<0.1–25)	(<0.1–24)	(<0.1–69)
	Cuttlefish	9.5	15	6.7	32	6.2	6.4	4.8	17
	Fish	7.7	10	6.6	25	4.6	4.4	2.5	12
		(2.9–12)	(4.0–18)	(2.2–15)	(9.1–45)	(<0.1–9.8)	(<0.1–7.8)	(<0.1–11)	(<0.1–28)
	Mollusc	7.3	7.7	11	26	<0.1	2.6	2.6	5.2
		(5.8–8.7)	(5.9–9.6)	(5.5–18)	(17–36)	(<0.1)	(<0.1–5.1)	(<0.1–5.2)	(<0.1–10)
	Sea cucumber	5.4	9.0	18	32	3.8	3.8	0.3	7.9
		(3.7–14)	(4.5–28)	(<0.1–110)	(8.2–151)	(<0.1–25)	(<0.1–9.8)	(<0.1–3.5)	(<0.1–39)
	Sea star	6.8	7.7	8.0	23	2.9	4.8	3.8	12
		(5.3–9.6)	(5.5–11)	(3.0–18)	(14–39)	(<0.1–5.0)	(4.3–5.3)	(<0.1–9.6)	(4.3–20)
	Shrimp	7.3	11	29	47	2.1	3.9	<0.1	6.0
		(5.9–8.2)	(7.9–17)	(4.6–69)	(18–93)	(<0.1–4.3)	(2.3–4.8)	(<0.1)	(2.3–9.1)
	Snail	6.6	15	24	46	1.2	4.2	2.0	7.3
		(4.1–9.8)	(6.9–39)	(3.6–54)	(15–103)	(<0.1–4.1)	(<0.1–6.8)	(<0.1–4.7)	(<0.1–16)
Tinggi Island	Clam	8.6	18	17	44	9.2	4.4	5.4	19
		(6.1–10)	(7.2–43)	(4.7–44)	(18–98)	(<0.1–31)	(<0.1–8.2)	(<0.1–13)	(<0.1–53)
	Crab	8.5	11	13	33	16	6.2	1.7	25
		(3.9–14)	(5.0–18)	(6.2–30)	(15–62)	(<0.1–61)	(2.5–9.8)	(<0.1–8.7)	(2.5–80)
	Cuttlefish	8.7	12	9.2	29	6.3	6.8	5.0	8.0
		(8.6–8.8)	(11–12)	(8.4–10)	(28–31)	(6.1–6.4)	(6.5–7.0)	(5.0)	(18–19)
	Fish	8.8	12	7.5	28	5.6	6.1	4.0	16
		(5.0–21)	(6.8–18)	(3.6–17)	(15–56)	(<0.1–10)	(3.5–8.9)	(<0.1–17)	(4.0–36)
	Sea cucumber	5.4	8.0	8.2	22	3.8	4.3	1.1	9.2
		(<0.1–9.1)	(3.0–12)	(<0.1–24)	(3.0–45)	(<0.1–6.7)	(<0.1–7.4)	(<0.1–5.6)	(<0.1–20)
	Sea star	7.6	11	5.6	24	2.6	6.2	8.0	17
		(6.7–8.4)	(9.4–12)	(5.5–5.8)	(22–26)	(<0.1–5.2)	(5.5–6.8)	(<0.1–16)	(6.0–28)
	Sea urchin	5.8	6.6	3.4	16	<0.1	4.7	<0.1	1.7
	Shrimp	8.6	21	12	42	17	4.6	<0.1	22

Parentheses show the range of concentration of each compound.

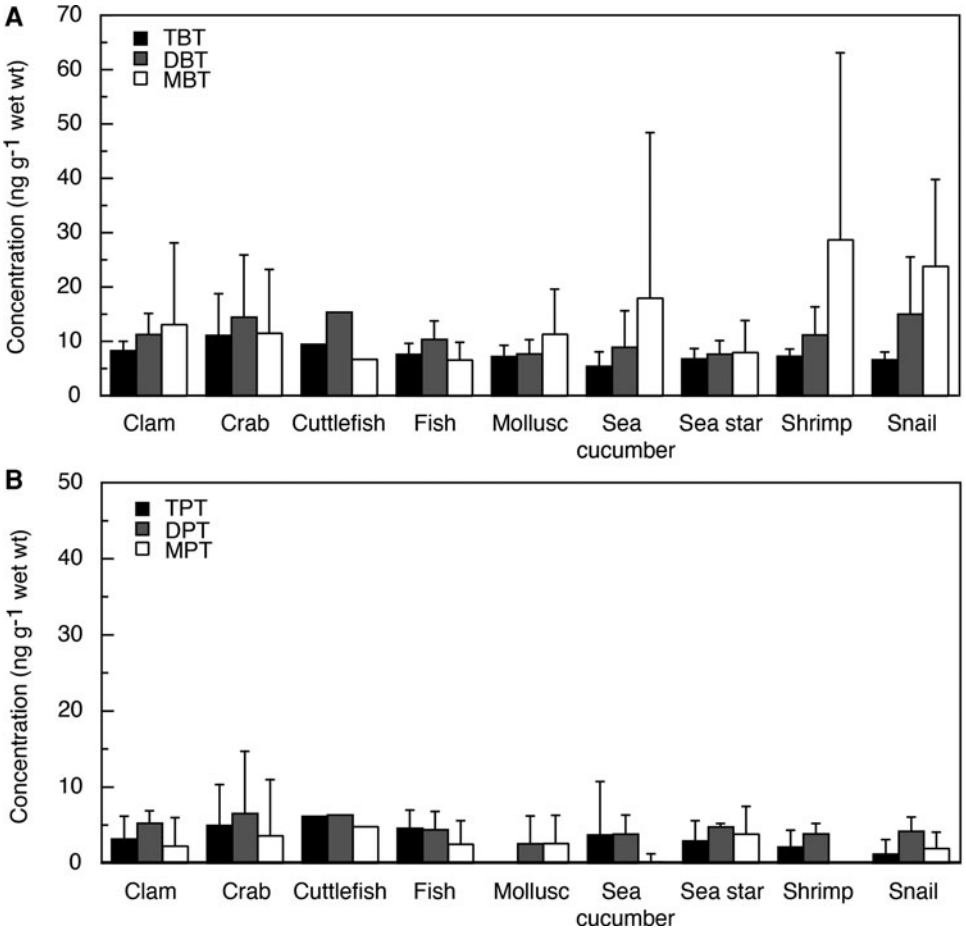

Fig. 2. Concentrations of BTs (A) and PTs (B) in biota from Merambong.

mean concentrations of TBT among each species of aquatic organisms were similar to each other. In most aquatic organisms from Tinggi Island, the ratios of DBT and MBT among BTs were higher than those of TBT (Figure 5).

The concentrations of PTs in each species of aquatic organisms from Merambong and Tinggi Island were measured (Table 3, Figures 2 and 3). The mean concentrations of the total PTs (∑PTs = TPT + DPT + MPT) from Merambong and Tinggi Island were, respectively, 10.5 ng g^{-1} ww, in the range of <0.1–69 ng g^{-1} ww, and 15.1 ng g^{-1} ww, in the range of <0.1–88 ng g^{-1} ww. In both regions, mean concentrations of ∑PTs were significantly lower than those of BTs ($P < 0.0001$), and those concentrations from Tinggi Island were significantly higher than those from Merambong ($P < 0.0005$) (Table 3, Figures 2 and 3).

TPT was detected in the range of <0.1–25 ng g^{-1} ww in aquatic organisms from Merambong (Table 3, Figure 2). Although the rate of each PT varies among aquatic organisms from Merambong, the ratios of MPT and DPT were higher than those of TPT (Figure 4). TPT in aquatic organisms in Tinggi Island was detected in the range of <0.1–61 ng g^{-1} ww (Table 3, Figure 3). Although in clam, crab and shrimp TPT was the dominant compound among PTs, in the other aquatic organisms from Tinggi Island the rates of the PTs were similar to or the rates of degradation products of TPT were higher than that of TPT (Figure 5).

Comparison with OT concentrations among species of aquatic organisms

Mean concentrations of BTs and PTs in each species of aquatic organisms from Merambong and Tinggi Island are shown in Table 3, Figures 2 and 3. Mean concentrations of ∑BTs in each species of aquatic organisms from Merambong were highest in the order of shrimp > snail > crab > clam > sea cucumber > cuttlefish > mollusc > fish > sea star. The concentrations of ∑PTs were highest in the order of cuttlefish > crab > sea star > fish > clam > sea cucumber > snail > shrimp > mollusc. The average concentration of ∑BTs in each species of aquatic organisms from Tinggi Island was highest in the order of clam > crab > shrimp > cuttlefish > fish > sea star > sea cucumber > sea urchin. The average concentrations of ∑PTs were highest in the order of crab > clam > cuttlefish > shrimp > sea star > fish > sea cucumber > sea urchin. Mean concentrations of BTs were higher than those of PTs in the samples from Tinggi Island, similar to the pattern in the samples from Merambong as shown in Table 3, Figures 2 and 3.

The relationships between δ^{15}N and each OT concentrations in the aquatic organisms were calculated to elucidate these factors. No correlation was observed between δ^{15}N and BT or PT in Merambong ($P > 0.05$) (Table 4). In contrast, although the correlation coefficients between δ^{15}N and OT compounds in aquatic organisms from Tinggi Island were low, positive correlations were observed between δ^{15}N on the one hand and, on the other, DBT, DPT, TPT and ∑PTs ($P < 0.05$) (Table 5).

The relationships between weight and OT concentrations in aquatic organisms were also investigated (Table 6). Negative correlations between weight and ∑BTs were also observed in fish from Merambong ($P < 0.05$), and negative correlations between weight and ∑PTs were observed in clam from Tinggi Island ($P < 0.05$). However, no correlations between weights and OT concentrations in the other aquatic organisms were observed ($P > 0.05$).

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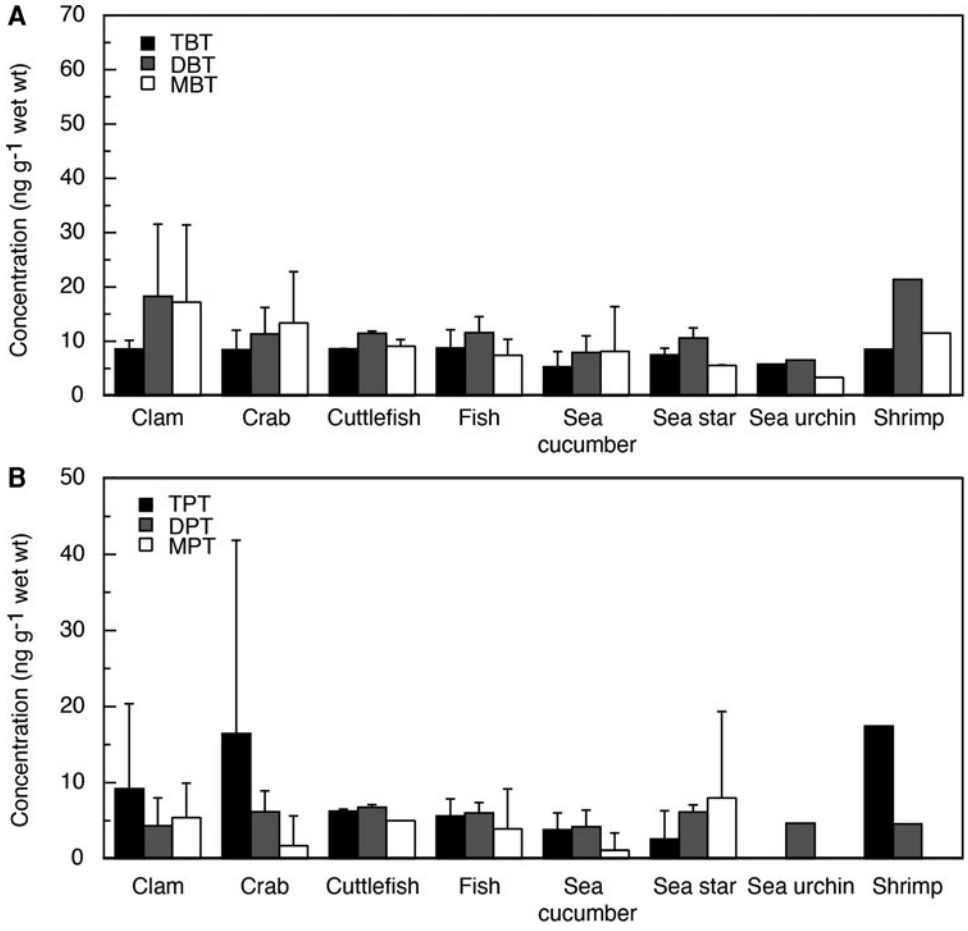

Fig. 3. Concentrations of BTs (A) and PTs (B) in biota from Tinggi Island.

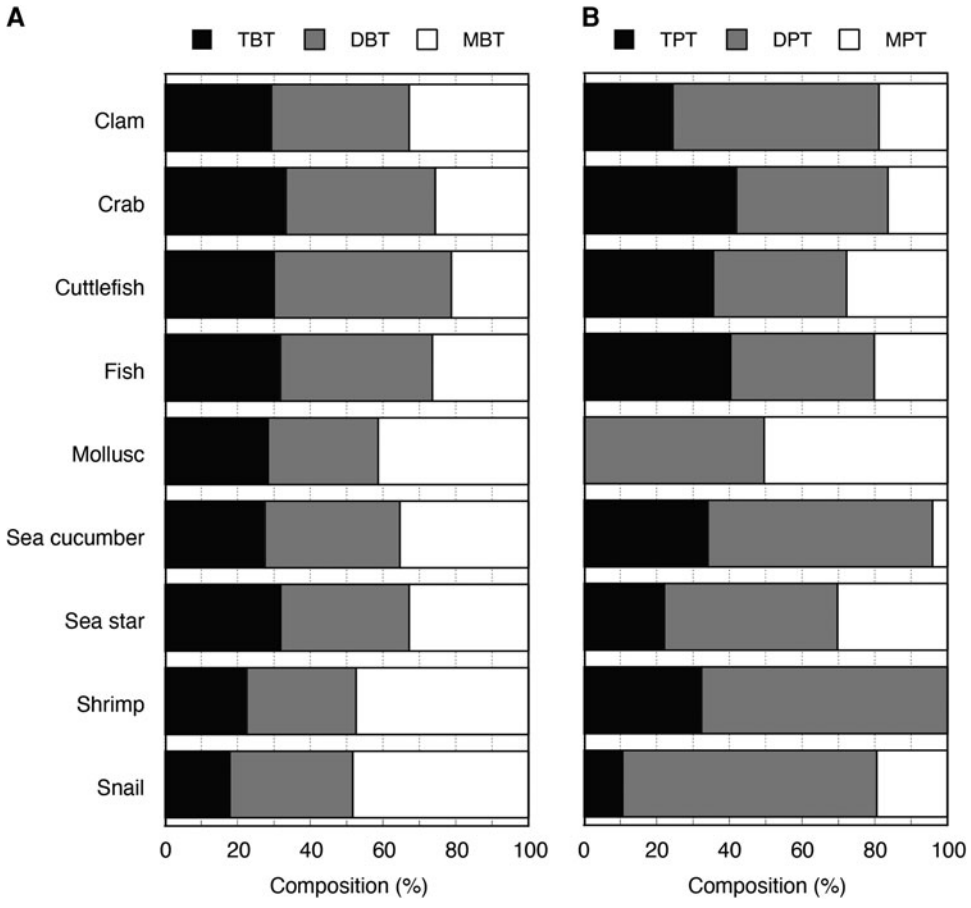

Fig. 4. Compositions of BTs (A) and PTs (B) in biota from Merambong.

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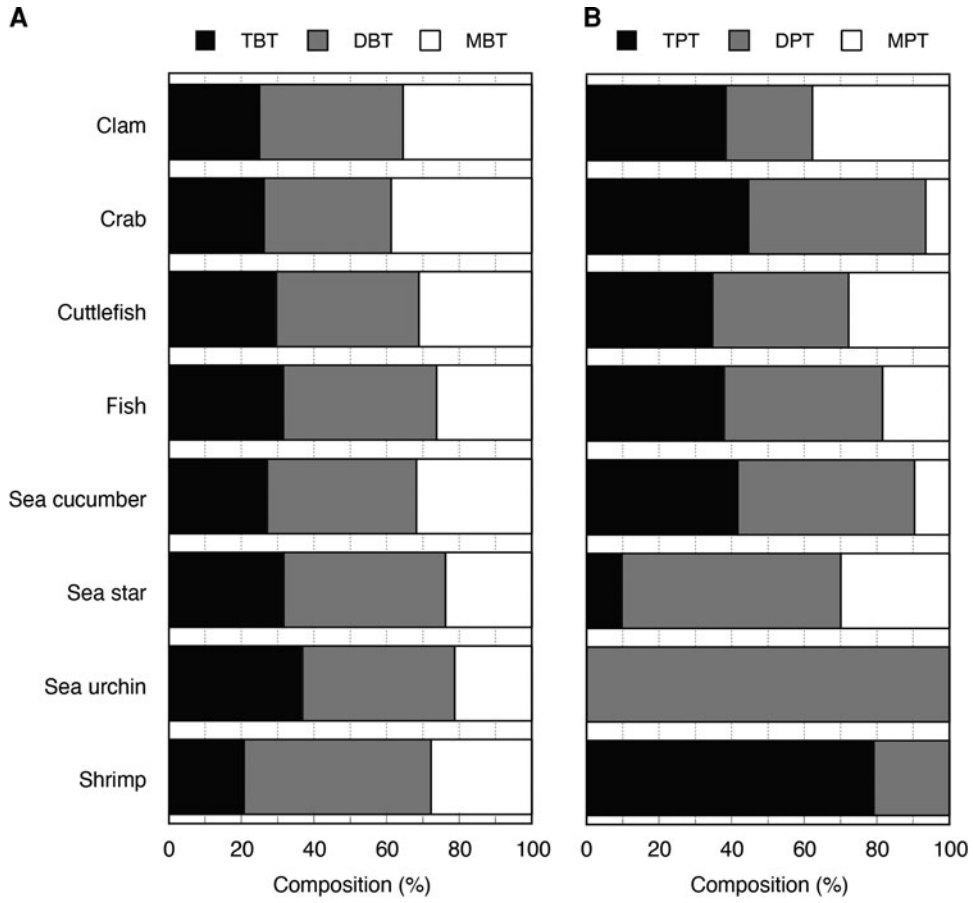

Fig. 5. Compositions of BTs (A) and PTs (B) in biota from Tinggi Island.

Table 4. Correlation coefficient between each organotin compound and δ^{15}N in biota from Merambong

	δ15N (‰)	MBT	DBT	TBT	MPT	DPT	TPT	ΣBTs	ΣPTs
δ^{15}N (‰)	1.000								
MBT	−0.141	1.000							
DBT	−0.012	0.622**	1.000						
TBT	−0.017	0.427**	0.790*	1.000					
MPT	0.099	0.274	0.311*	0.226	1.000				
DPT	0.098	0.218	0.502**	0.636**	0.180	1.000			
TPT	0.184	0.222	0.431**	0.542**	0.160	0.585**	1.000		
ΣBTs	−0.112	0.912**	0.834**	0.659**	0.316*	0.376**	0.342*	1.000	
ΣPTs	0.209	0.344*	0.559**	0.589**	0.624**	0.749**	0.738**	0.467**	1.000

*P < 0.05, **P < 0.01.

Table 5. Correlation coefficient between each organotin compound and δ^{15}N in biota from Tinggi Island

	δ15N (‰)	MBT	DBT	TBT	MPT	DPT	TPT	ΣBTs	ΣPTs
δ^{15}N (‰)	1.000								
MBT	0.057	1.000							
DBT	0.282*	0.607**	1.000						
TBT	0.247	0.455**	0.794**	1.000					
MPT	0.239	0.162	0.208	0.244	1.000				
DPT	0.339*	0.328*	0.733**	0.774**	0.274	1.000			
TPT	0.337*	0.264	0.623**	0.667**	0.197	0.703**	1.000		
ΣBTs	0.111	0.919*	0.79**	0.67**	0.131	0.495**	0.395**	1.000	
ΣPTs	0.297*	0.176	0.519**	0.556**	0.619**	0.596**	0.819**	0.271	1.000

*P < 0.05, **P < 0.01.

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Table 6. Correlation coefficient between weight and ΣBTs or ΣPTs in biota from Merambong and Tinggi Island

	Clam		Crab		Fish		Sea cucumber		Sea star	Snail
	Merambong	Tinggi Island	Merambong	Tinggi Island	Merambong	Tinggi island	Merambong	Tinggi Island	Merambong	Merambong
ΣBTs	−0.321	0.369	0.179	−0.359	−0.410*	−0.248	0.014	0.192	−0.154	−0.634
ΣPTs	0.643	−0.741*	−0.459	−0.821	0.028	−0.295	−0.163	−0.055	0.564	−0.415

*$P < 0.05$.

Concentrations of organotin compounds in the tissues and organs of fish from seagrass area of Merambong

The concentrations of BTs were measured in muscle, liver and egg of fish from Merambong (Figure 6). BTs were detected in all tissues and organs examined in the present study. The mean concentrations of ∑BTs (24.5 ± 23.0 ng g^{-1}) in liver were significantly higher than those in muscle (7.1 ± 1.9 ng g^{-1}) ($P < 0.01$). The level of ∑BTs in egg was similar to that in muscle. In contrast, PTs were detected in few samples. No significant differences in ∑PTs were observed between muscle and liver ($P > 0.1$), and those levels were similar to those in egg.

The rates of TBT, DBT and MBT among BTs are shown in Figure 7. In most fish, MBT was dominant among BTs, followed by DBT. In *Eleuteronema tetradactylum*, the rates of TBT in muscle and egg were higher than that in liver. TPT accounted for 100% of the total PTs in muscle, liver and egg, although MPT accounted for 100% in one muscle sample.

Discussion

OT concentrations in aquatic organisms

Butyltin compounds were detected in aquatic organisms from Merambong and Tinggi Island. After the ban of the use of TBT by the IMO, there have been few reports on the concentrations of BT compounds in biota. For example, the concentrations of BTs were 16–31 ng g^{-1} dw in banded murex *Hexaplex trunculus* from the Adriatic Sea (Cacciatore *et al.*, 2018), 33–73 ng g^{-1} dw in rock shell *Reisha clavigera* from Hong Kong (Ho & Leung, 2016) and 34–1676 ng g^{-1} dw in mussel *Mytilus galloprovincialis* from the Croatian Adriatic Coast (Furdek *et al.*, 2012). These concentrations of BTs in biota after the ban on OTs were in the range of 16–1676 ng g^{-1} dw. However, these values were on a dry weight basis. To compare with the concentrations of BTs and PTs, it is necessary to convert these values into wet weight. Verhaegen *et al.* (2012) reported that the dry weight to wet weight ratio of peeled shrimp was 22.4 ± 1.1%. Therefore, the mean water content of aquatic organisms was assumed to be 80%. Converted to a wet weight basis, the reported concentrations of BTs in aquatic organisms after the ban of OTs were in the range of 3.2–335 ng g^{-1} ww. The concentrations of BTs from Merambong and Tinggi Island were in the range of 8.2–151 ng g^{-1} ww and 3–109 ng g^{-1} ww, respectively. The concentrations of BTs from both locations were similar to the reported values.

TBT detected in samples from Merambong and Tinggi Island were in the range of 2.9–28 ng g^{-1} ww and <0.1–21 ng g^{-1} ww, respectively. TBT concentrations were 4–124 ng g^{-1} dw in brown shrimp (*Crangon crangon*) from the North Sea (Verhaegen *et al.*, 2012), <4–18 ng g^{-1} dw in *H. trunculus* from the Adriatic Sea (Cacciatore *et al.*, 2018), 13.3–35.7 ng g^{-1} dw in *R. clavigera* from Hong Kong (Ho & Leung, 2016) and <6–1045 ng g^{-1} dw in *M. galloprovincialis* from the Croatian Adriatic Coast (Furdek *et al.*, 2012). When converted to a wet weight basis assuming a water content of 80%, the concentrations of TBT in aquatic organisms after the ban on OTs were in the range of <1.2–209 ng g^{-1} ww. TBT concentrations in samples from Merambong and Tinggi Island were near the low ends of the ranges of reported values.

Although the concentrations of TBT in aquatic organisms from Merambong and Tinggi Island were similar among the species, the ratios of TBT degradation products DBT and MBT among the total BTs were higher than those of TBT. Ohji *et al.* (2019) reported that the rates of degradation products in sediment and leaves collected from Merambong and Tinggi Island in 2012 were higher than those of TBT. The rate of each BT in the aquatic

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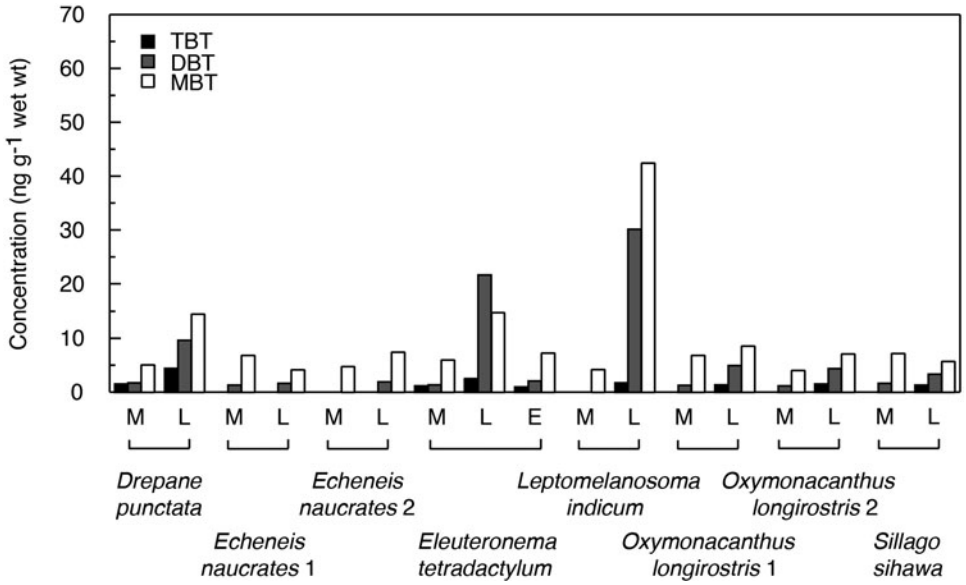

Fig. 6. Concentrations of BTs in muscle, liver, and egg of fish in seagrass area of Merambong, 2013. M, muscle; L, liver; E, egg.

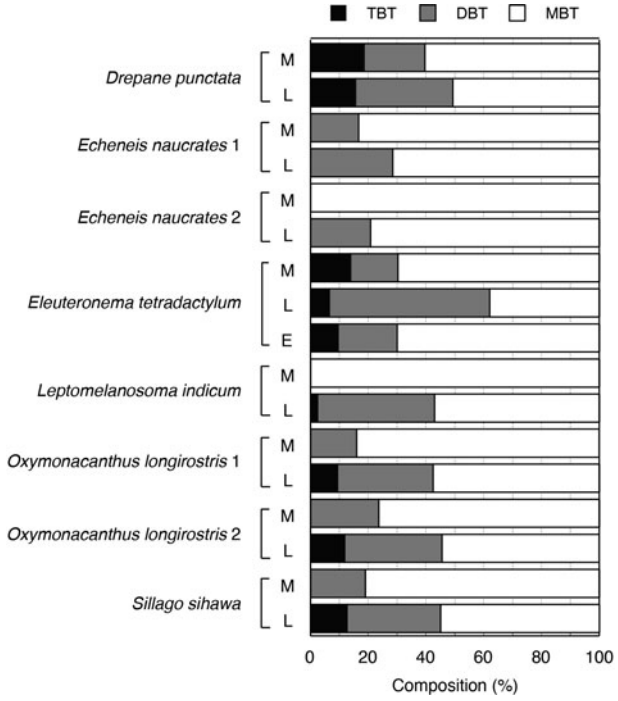

Fig. 7. Compositions of BTs in fish from seagrass area of Merambong, 2013. M, muscle; L, liver; E, egg.

organisms in this study showed a similar trend to that of sediment and leaves.

No significant differences in TBT concentrations between Merambong and Tinggi Island were observed in this study. Ohji *et al.* (2019) reported that the average concentrations of TBT in sediment from Merambong and Tinggi Island in 2012 were 4.5 ng g^{-1} dw and 4.2 ng g^{-1} dw, respectively, and that TBT concentrations in Merambong were similar to those in Tinggi Island. Moreover, no differences were observed in average concentrations of BTs in mangrove leaves between those two regions. The horizontal distributions of TBT concentrations in aquatic organisms were similar to those in sediment and leaves.

The concentrations of BTs in aquatic organisms from Merambong were similar to those from Tinggi Island. Ohji *et al.* (2019) reported that the BT concentrations in sediment from Merambong were higher than those in sediment from Tinggi Island in 2012, and there were no significant differences in the total BT levels in leaves between two regions. The BT concentrations in sediment show contamination from the past, because the half-lives of BTs in sediment are several years (Langston *et al.*, 2015). On the other hand, the concentrations of BTs in leaves show current contamination. In aquatic organisms, the trend of BT concentrations between Merambong and Tinggi Island was similar to that in leaves; Merambong is located in an industrial area, and Tinggi Island is used as a resort area. Despite the differences in industrial bases the status of BT contamination in aquatic organisms is similar between two locations.

Phenyltin compounds were also detected in aquatic organisms from Merambong and Tinggi Island. There were few reports regarding the levels of PTs in biota. ∑PT concentrations were 541–6855 ng g^{-1} dw in *R. clavigera* from Hong Kong (Ho & Leung, 2016), and <11–126 ng g^{-1} dw in *M. galloprovincialis* from the Croatian Adriatic Coast (Furdek *et al.*, 2012). These concentrations of PTs in biota after the ban on OTs were in the range of <11–6855 ng g^{-1} dw. Converting those concentrations to a wet weight basis, assuming water content of 80%, the concentrations of PTs in biota after the ban were in the range of <2.2–1371 ng g^{-1} ww. The concentrations of PTs from Merambong and Tinggi Island were in the ranges of <0.1–69 ng g^{-1} ww and <0.1–88 ng g^{-1} ww, respectively. PT concentrations in Merambong and Tinggi Island were near the low ends of the ranges of the reported values, as were BT concentrations.

TPT was detected in each species from Merambong and Tinggi Island in the ranges of <0.1–25 ng g^{-1} ww and <0.1–61 ng g^{-1}, respectively. TPT levels were 1–24 ng g^{-1} dw in *C. crangon* from the North Sea (Verhaegen *et al.*, 2012), 489–6404 ng g^{-1} dw in *R. clavigera* from Hong Kong (Ho & Leung, 2016) and <11–126 ng g^{-1} dw. in *M. galloprovincialis* from the Croatian Adriatic Coast (Furdek *et al.*, 2012). Conversion of these reported values to a wet weight basis, assuming 80% water content, gives us 0.2–1281 ng g^{-1} ww of TPT in biota after the ban on OTs. Triphenyltin concentrations in Merambong and Tinggi Island were near the low ends of the ranges of reported values.

The concentrations of total PTs in aquatic organisms from Tinggi Island were significantly higher than those from

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Merambong. Ohji *et al.* (2019) did not detect PTs in sediment and did not observe differences in PT concentrations in leaves between the two areas in 2012. TPT in aquatic organisms from Merambong and Tinggi Island were detected in the range of <0.1–25 ng g^{-1} ww and <0.1–61 ng g^{-1}, respectively. Ohji *et al.* (2019) observed no differences in TPT levels between the two areas in 2012. The horizontal distributions of total BT and TPT in aquatic organisms were different from those in leaves.

Comparison of OTs concentrations among species of aquatic organisms

The differences in BT and PT concentrations in species of aquatic organisms are shown in Figures 2 and 3. The concentrations were similar between the two locations. Namely, the concentrations of BTs and PTs in crab and clam were high and those in fish were low. Crab and clam have narrow ranges of activity and are deposit feeders. Fish have a wide range of activity, and their food is mainly plankton and algae. In other words, the difference in concentrations may be due to the respective diets and habitats of these animals.

The correlations between δ^{15}N and OTs are discussed, because δ^{15}N was used as a food web indicator (Tables 4 and 5). Although weak, correlations were found between δ^{15}N on the one hand and, on the other, each TPT as well as total PT concentrations in biota from Tinggi Island, whereas no correlations were found between δ^{15}N and each TBT and total BT concentrations. It was reported that TBT was not accumulated through the food web (Stäb *et al.*, 1996; Kono *et al.*, 2008). On the other hand, Kono *et al.* (2008) reported that TPT was accumulated through the food web. As these reports were published before the ban on TBT and TPT by the IMO, it is considered that these trends occurred when there were many TBT and TPT loads to the aquatic environment. Meanwhile, no correlation was observed between δ^{15}N and each OT concentration in biota from Merambong. It is considered that TBT and TPT were not accumulated through the food web, because the loads of TBT and TPT have decreased.

Correlations were found between TBT and TPT or total PT in Merambong and Tinggi Island. This suggested that TPT was used as an antifouling paint as well as TBT, and that, despite the small scale of the loads to the aquatic organisms, it is presumed that the accumulations of TBT and TPT in aquatic organisms have continued. It was reported that an extremely low concentration of TBT is toxic to susceptible organisms, such as molluscs (Gibbs *et al.*, 1988; Lee, 1996) and crustaceans (Ohji *et al.*, 2002, 2003), which might lead to disturbances in the dynamics of the populations in ecosystems.

Table 6 shows the relationships between the weights of various aquatic organisms and total BT or total PT. The correlation coefficient of the weight of fish from Merambong and total BTs, and that of the weight of clam from Tinggi Island and total PTs, were −0.410 and −0.741, respectively. It is considered that the correlation coefficient became negative because the loads of these OTs decreased, and that OT concentrations were diluted by the growth of aquatic organisms. No correlations were found between the weight of each species of the other aquatic organisms and the BT or PT concentration.

Concentrations of organotin compounds in the tissues and organs of fish from Merambong

The concentrations of BT compounds in muscles, liver and eggs of fish are shown in Figure 6. The BT concentrations in liver were higher than those in muscle. The rate of MBT was higher than those of the other BT compounds (Figure 7). These results shows that TBT absorbed by fish is concentrated in the liver and degraded. Among BTs, DBT and MBT were detected at high levels in the liver tissue of the pike (Stäb *et al.*, 1996). TBT is known to be dealkylated in the liver by ethoxyresorufin *O*-deethylase (EROD) activity and P-450 enzymes (Fent & Bucheli, 1994), thus in the present study TBT was degraded and excreted from fish via bile. The BT concentration in egg in *Eleuterronea letradactylum* is also shown in Figure 6. Considering the transition of BTs to the next generation, it is important to measure BT concentrations in eggs. The BT concentrations in eggs were of similar levels to those in muscle, suggesting the transition of BTs from parental females to their eggs. It was also reported that BTs transfer from parental females to offspring in the viviparous surfperch *Ditrema temmincki* (Ohji *et al.*, 2018). BTs are suspected of having the potential for deleterious effects in the eggs or offspring of fish such as *E. tetradactylum* and *D. temmincki*. TBT has adverse effects in fish, such as physiological abnormality even at ambient water levels (e.g. Hall & Bushong, 1996). Therefore, these contaminants might affect these fishes in the coastal ecosystems.

Conclusion

BT and PT concentrations were measured in various species of aquatic organisms from Merambong and Tinggi Island, Malaysia and were found to be within the reported ranges. The two regions showed no differences in levels of BT contamination. Among BTs, the ratios of DBT and MBT, which are the degradation products of TBT, were higher than those of TBT. The concentrations of PTs in Merambong and Tinggi Island were within the reported ranges. No differences were observed in TPT levels between the two regions.

No correlation was found between δ^{15}N and the concentrations of each OT in biota from Merambong, suggesting that TBT and TPT do not accumulate through the food web. However, it is presumed that the intake of both TBT and TPT in aquatic organisms has decreased, because weight was negatively correlated with TBT concentrations in aquatic organisms. In tissues and organs of fish, BT concentrations were higher in liver than in muscle. It is noteworthy that BTs were detected in egg, implying that there is a potential for BTs to affect future generations.

Thus, even after OTs were banned, they were detected in aquatic organisms, although at lower concentrations. As the present OTs levels still have the potential to affect marine organisms, it is important to continue the monitoring of OTs in biota.

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