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Organotin Contaminations in Malaysia

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Abstract—The current status of contaminations by organotin (OT) compounds in Malaysia was reviewed. Tributyltin (TBT) in water samples ranged from $<1.39\,\mathrm{ng\,L^{-1}}$ to $115\,\mathrm{ng\,L^{-1}}$. The higher concentrations of TBTs in water samples were observed in mooring facility of pleasure crafts and the Strait of Malacca with heavy shipping traffic. TBT to total butyltin (BT) was dominant species in water samples, suggesting the latest use of TBT. Phenyltin (PT) compounds were not detected in water samples. Tributyltin in sediment samples ranged from $<0.3\,\mu\mathrm{g\,kg^{-1}}$ dry to $492\,\mu\mathrm{g\,kg^{-1}}$ dry. TBT concentrations in sediment samples were high in the Strait of Johor where locates between Malaysia and Singapore. In the case of sediment, the higher percentage of degradation products of TBT was observed among total BTs. The concentrations of triphenyltin (TPT) compounds in sediment ranged from $0.1-34\,\mu\mathrm{g\,kg^{-1}}$ dry. TBT in bivalves and fish also detected at the range of $<0.5-299\,\mu\mathrm{g\,kg^{-1}}$, $2.4-190\,\mu\mathrm{g\,kg^{-1}}$, respectively. TBT concentrations in some biological samples exceeded the tolerable average residue levels (TALR) which calculated from tolerable daily intake (TDI) for TBT. TBT was not detected in biological samples.

Most gastropods from Malaysia were caused imposex. Especially, the highest incidence was found in the Southern part of the Strait of Malacca. Geographical distribution of incidence of imposex accorded with that that of concentration of TBT in aquatic environment.

Key words: butyltin, phenyltin, Malaysia, concentrations, human risk

Introduction

Malaysia where locates in central parts of Southeast Asia, consists of two geographical regions of Peninsular Malaysia and Malaysian Borneo divided by the South China Sea. In the last two decades, Malaysia has undergone tremendous growth and prosperity, because Malaysia has an abundance of mineral resource and is area richly endowed with nature. As the consequence, tourist and trade business were activated and the Strait of Malacca and the Strait of Johor in Malaysia became the world's busiest waterways. The development of economic activity has a potential of marine contamination by chemical substances. Monirith et al. (2003) reported that polychorinated biphenyls (PCBs), DDT and its metabolites, hexachlorocyclohexane (HCH), Chlordane (CHLs) and hexachlorobenzene (HCB) in all mussel samples from Malaysia were detected. Sudaryant et al. (2005) reported that persistent organochlorine compounds (POPs) such as polychlorinated dibenzo-p-dioxins and dibenzofurans, PCBs, organochrorine pesticide and tris(4-chlorophenyl) methane were detected in human breast milk. These reports show the seriousness of the chemical pollution in Malaysia.

Antifouling biocides were developed to prevent the attachment of marine organisms on the hull of ship. Organotin (OT) compounds as among antifouling biocides were the effective chemical substance. However, OTs which were leached into water from boat hulls have caused the imposex for dog-whelk (*Nucella lapillus*) and the malformation for Pacific oyster (*Crassostrea gigas*) (Alizieu et al. 1986, Gibbs and Bryan 1986). As the consequence, aquatic resource has been decreased. In the developed countries such as England, USA, Canada etc., the use of tributyltin (TBT) compounds has been regulated around 1980, for instance, establishment of environmental quality target, control of release rates from antifouling paint and ban on the use of TBT-containing antifouling paints on pleasure crafts less than 25 m length. On the other hand, the use of TBT based antifouling paints in developing countries is not restricted even now. Therefore, OTs contaminations in Southeast Asia which is developing country attract attention recent year. Many studies such as mussel watch revealed the wide spread contamination by OTs in

Table 1. The concentration of organotin compounds in water/ng Sn L^{-1} All values are converted to Sn ions. Parentheses are mean values.

Year	Location	MBT	DBT	TBT	Ref.
1991–1992	Selangor	–	–	<1.39–115	Tong et al., 1996
		–	–	(21)	Tong et al., 1996
	Negeri Sembilan	–	–	<1.39–20	Tong et al., 1996
		–	–	(10)	Tong et al., 1996
	Terengganu	–	–	3.9–25	Tong et al., 1996
			–	(12)	Tong et al., 1996
1996	Strait of Malacca	0.1–2.4	<0.04–0.8	0.1–1.9	Hashimoto et al., l998
		(0.72)	(0.5)	(0.79)	Hashimoto et al., 1998
	Bay of Bengal	<0.04–0.1	<0.04	<0.04–0.1	Hashimoto et al., 1998
	(Open Sea)	(<0.04)	(<0.04)	(<0.04)	Hashimoto et al., 1998

Southeast Asia (Sudaryanto et al. 2002, Midorikawa et al. 2004a,b, Harino et al. 2006). Higher concentrations of butyltin (BT) were found in mussels collected from the locations with intensive maritime activities and aquaculture areas in Malaysia. Input of OTs will decrease in near future, because the usage of OTs was banned in IMO. It is important to evaluate how much this treaty helps for recovery of marine environment of the Malaysia. To do that, the comparison of the appearance of OTs contamination between presence and future are needed. In this paper, the current status of OTs contaminations is reviewed.

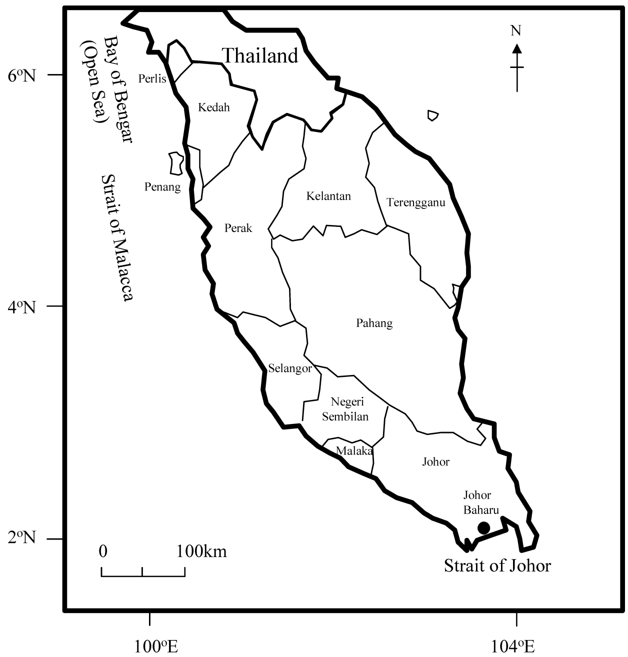

Fig. 1. The map of Malaysia.

Concentrations of organotin compounds in water

There is a few available data concerning OTs concentration in water column from Malaysia (Table 1). Tong et al. (1996) measured the concentration of TBT in 26 sites of Peninsular Malaysia in 1991–1992. The concentrations of TBT were in the range of <1.39–115 ng L^{-1}. Maximum concentration of TBT was observed in the docking site of a yacht club where many pleasure crafts were moored. TBT concentrations were subsequently high in the sites of a mussel/aquaculture farm. The lowest levels of TBT found at the wider channel to North Port. Hashimoto et al. (1998) surveyed the BTs along the Strait of Malacca (Fig.1). TBT concentrations in the Strait of Malacca were higher than those in Open Ocean (Bay of Bengal). and were similar to in Tokyo Bay, where is heavy shipping traffic. TBT and total BTs concentrations in the seawater samples decreased with increasing distance from Singapore along the Strait of Malacca. This suggests that the main source of the contamination by BTs is large vessels in Singapore. Furthermore, the higher ratio of TBT/DBT was observed in the Strait of Mallacca, suggesting the recent inputs of TBT.

Hashimoto et al. (1998) also reported that phenyltin (PT) compounds were not detected in water samples along the Strait of Malacca and Open Sea (Bay of Bengal).

Concentrations of organotin compounds in sediment

The concentrations of OTs in sediment from Malaysia were firstly reported in 1996 by Tong et al. (1996). TBT concentrations in sediment from Selangor, Negeri Sembilan and Terengganu were ranged from <0.3–89 μg kg^{-1} dry (Fig.1, Table 2). Maximum concentrations of TBT were observed in port area from Terengganu and TBT concentrations in sediment from Tanjung Tuan, Port Dickson of Negeri Sembilan where are a recreational beach area were under of detection limit. Sudaryanto et al. (2004) reported that TBT concentrations in sediment from the coastal area in Malaysia were ranged of 1.1–94 μg kg^{-1} dry. Sediment samples were taken from areas such as recreational beach, small fishing boats and small scale aquaculture revealed lower concentration of TBT. While, consistently elevated concentrations of BTs were found along the coast of the Strait of Malacca, particularly in locations having intensive maritime activities such as Penang, Johor and Johor Bahru (Fig. 1). Although the proportions of degradation products (dibutyltin (DBT) plus monobutyltin (MBT)) in total BTs general accounted for

Table 2. The concentration of organotin compounds in sediment/µg Sn kg^{-1} dry All values are converted to Sn ions. Parentheses are mean values. * : µg Sn kg^{-1} wet

Year	Locations	MBT	DBT	TBT	MPT	DPT	TPT	Ref.
1991–1992*	Selangor	–	–	12	–	–	–	Tong et al., 1996
	Negeri Sembilan	–	–	<0.3–11	–	–	–	Tong et al., 1996
		–	–	(5.7)	–	–	–	Tong et al., 1996
	Terengganu	–	–	89	–	–	–	Tong et al., 1996
1997–1998	Langkawi	6.0–8.1	2.2–2.9	1.1–2.0	–	–	–	Sudayyanto et al., 2004
		(7.1)	(2.5)	(1.6)	–	–	–	Sudayyanto et al., 2004
	Kedah	3.4	1.9	2.1	–	–	–	Sudayyanto et al., 2004
		(3.4)	(1.9)	(2.1)	–	–	–	Sudayyanto et al., 2004
	Penang	17–53	9.2–76	12–94	–	–	–	Sudayyanto et al., 2004
		(35)	(43)	(53)	–	–	–	Sudayyanto et al., 2004
	Selangor	4.1	2.6	6.5	–	–	–	Sudayyanto et al., 2004
	Negeri Sembilan	11	3.0	2.3	–	–	–	Sudayyanto et al., 2004
	Melaka	14	8.7	53	–	–	–	Sudayyanto et al., 2004
	Johore	26	13	17	–	–	–	Sudayyanto et al., 2004
	Johore Bahru	15–243	6.1–158	11–450	–	–	–	Sudayyanto et al., 2004
		(104)	(63)	(156)	–	–	–	Sudayyanto et al., 2004
2006	Langkawi	19	7.1–8.8	15–19	16–32	0.4–11	0.2–13	Harino et al., 2008
		(19)	(8.0)	(17)	(24)	(5.4)	(6.8)	Harino et al., 2008
	Perlis	17	7.9	8.9	77	24	28	Harino et al., 2008
	Penang	5.5–24	1.4–48	0.9–20	<0.1–30	0.7–8.2	0.6–11	Harino et al., 2008
		(14)	(5.3)	(10)	(18)	(3.4)	(4.1)	Harino et al., 2008
	Perak	5.3–14	1.3-4.7	1.5–1.9	<0.1–81	0.4–19	0.1–24	Harino et al., 2008
		(9.6)	(3.0)	(1.7)	(41)	(9.6)	(12)	Harino et al., 2008
	Negeri Sembilan	4.1–12	1.1–2.5	0.7–1.1	<0.1–44	0.5–17	0.2–22	Harino et al., 2008
		(8.0)	(1.8)	(0.9)	(22)	(8.7)	(11)	Harino et al., 2008
	Johor	25–241	13–186	41–228	<0.1–121	0.7–27	0.4–34	Harino et al., 2008
		(89)	(61)	(144)	(46)	(8.4)	(9.0)	Harino et al., 2008
	Strait of Johor	82–542	30–232	41–492	34–68	5.1–29	0.3–34	Harino et al., 2008
		(339)	(143)	(215)	(53)	(14)	(12)	Harino et al., 2008

58–83% in sediment from Malaysia, higher proportions of TBT in sediment (50–79%) were observed in areas of high maritime activities, such as Penang Bride, Port Klang and Johor Bahru. Harino et al. (2007) reported the latest data of OTs. The concentrations of MBT, DBT and TBT in sediment from the coastal water of Peninsular Malaysia were in the range of 4–242 µg kg^{-1} dry, 1–186 µg kg^{-1} dry and 0.7–228 µg kg^{-1} dry, respectively. The higher concentrations of TBT were observed in the southern part of Peninsular Malaysia and the ratios of TBT among BTs were also higher in this part, showing the higher input of TBT in southern part of Malaysia. The concentrations of MBT, DBT and TBT in sediment from the coastal water of the Strait of Johor were in the range of 83–542 µg kg^{-1} dry, 30–232 µg kg^{-1} dry and 41–492 µg kg^{-1} dry, respectively, which were higher than those in Peninsular Malaysia. In spite of higher concentration of TBT in comparison with those in Peninsular Malaysia, MBT was dominant compounds among BTs. It is suggested that the degradation rate of TBT is faster than the input rate in this area. The concentrations of monophenyltin (MPT), diphenyltin (DPT) and triphenyltin (TPT) were in the range of <0.1–121 µg kg^{-1} dry, 0.4–27 µg kg^{-1} dry and 0.1–34 µg kg^{-1} dry in sediment from the coastal water of Peninsular Malaysia, respectively and were in the range of 41–66 µg kg^{-1} dry, 5–29 µg kg^{-1} dry and 0.3–34 µg kg^{-1} dry from the Strait of Johor, respectively. The concentrations of PTs in Strait of Johor were similar to those in the coastal waters of Peninsular Malaysia. Monophenyltin (MPT) which is degradation compounds of TPT in sediment from Malaysia was dominant compounds among PTs.

The concentrations of OTs in sediment were compared among Malaysia, Thailand and Vietnam (Harino et al. 2007). The higher concentration and the wide variations of TBT and TPT in sediment from Malaysia were observed, showing that the more amounts of TBT have been used.

Concentrations of organotin compounds in biological samples

Mussel is an important item in order to grasp the current status of OT contaminations. Mussel watch program was carried out in various areas (Table 3). Tong et al. (1996) reported the concentration of TBT in cockles (*A. granosa*), soft-shell (*Paphia* sp.). Generally, TBT concentrations in these species were low at the range of <0.3–89 µg kg^{-1}.

Table 3. The concentration of organotin compounds in bivalves/µg Sn kg^{-1} All values are converted to Sn ions. Parentheses are mean values.

	Year	Locations	MBT	DBT	TBT	MPT	DPT	TPT	Ref.
Cockles	1991–1992	Selangor	–	–	<0.5–2.3	–	–	–	Tong et al., 1996
(*Anadara granosa*)			–	–	(1.3)	–	–	–	Tong et al., 1996
Soft-shelled calms		Selangor	–	–	1.5	–	–	–	Tong et al., 1996
(*Paphia* sp.)			–	–		–	–	–	Tong et al., 1996
Green mussels		Selangor	–	–	5.8–9.6	–	–	–	Tong et al., 1996
(*Perna viridis*)			–	–	(7.7)	–	–	–	Tong et al., 1996
Green Mussel	1997–1998	Sabah	<1.8–2.4	<0.5	1.4	–	–	–	Sudayyanto et al., 2002
(*Perna viridis*)			(1.5)	(0.2)	(1.4)	–	–	–	Sudayyanto et al., 2002
		Langkawi	4.4–11	2.1–3.1	4.5–45	–	–	–	Sudayyanto et al., 2002
			10	3.8	23	–	–	–	Sudayyanto et al., 2002
		Kedah	(3.8)	(1.2)	(9.0)	–	–	–	Sudayyanto et al., 2002
		Penang	21–39	10–13	61–106	–	–	–	Sudayyanto et al., 2002
			(30)	(11)	(84)	–	–	–	Sudayyanto et al., 2002
		Selangor	8.8	2	11	–	–	–	Sudayyanto et al., 2002
		Negeri Sembilan	<1.8–18	0.7–5.1	4.5–36	–	–	–	Sudayyanto et al., 2002
			(6.2)	(2.3)	(16)	–	–	–	Sudayyanto et al., 2002
		Melaka	5.2	1.2	6.1	–	–	–	Sudayyanto et al., 2002
		Johor	16	10	36	–	–	–	Sudayyanto et al., 2002
		Johor Bahru	6.3–50	4.2–82	20–299	–	–	–	Sudayyanto et al., 2002
			(27)	(36)	(125)	–	–	–	Sudayyanto et al., 2002
Green Mussel	2006	Langkawi	103	4	17	<1	<1	<1	Harino et al., 2008
(*Perna viridis*)		Perlis	53	2	4	8	1	<1	Harino et al., 2008
		Penang	88	4	32	22	5	<1	Harino et al., 2008
		Negeri Sembilan	77	3	8	<1	<1	<1	Harino et al., 2008
		Johor	42–132	3.5–20	11–59	<1–20	<1–3	<1	Harino et al., 2008
			(80)	(9)	(28)	(6)	(3)	(<1)	Harino et al., 2008

Sudaryanto et al. (2002) were reported the concentration of BTs in green mussels (*Perna viridis*) from 18 sites of the coastal area of Malaysia. The highest concentration of total BTs was found in mussels collected at the narrowest of the Strait of Johor as well as sediment samples. Sudaryanto et al. (2002) determined total tin in mussel samples and compared with the concentration between total BTs and total tin. A significant positive correlation was observed between the concentrations of total BTs and total tin. The highest total tin and BTs concentration was found in mussels from the narrowest of the Strait of Johor. Harino et al. (2007) reported that MBT, DBT and TBT compounds in green mussel (*Perna viridis*) samples from Malaysia were detected in the range of 41–102 µg kg^{-1}, 3–5 µg kg^{-1} and 8–32 µg kg^{-1}, respectively. Drastic difference of TBT concentration in mussel samples was not observed among sampling sites and MBT was dominant compounds in all sampling sites from Peninsular Malaysia. Concentrations of PTs were the values near the detection limit in most sampling sites from the Strait of Johor. MPT among PTs was dominant compounds in the Strait of Johor. Sudaryanto et al. (2002) carried out mussel watch in Asia Pacific areas. As the consequence, it was reported that mussel samples from Malaysia were contaminated by BTs heavily.

The concentrations of BTs in fish are shown in Table 4. TBT in fish was detected in the range of 2.4–190 µg kg^{-1}. The percentages of TBT were generally higher than its degradation products, DBT and MBT. Fish have greater metabolic capability than bivalves because of the presence of a high level of cytochrome P450 dependent mixed-function oxygenase enzymes. Despite this, high percentage of TBT suggests fresh input of TBT and the presence of contaminant sources along coastal waters of Malaysia. Among fish from Malaysia, the pelagic fish revealed higher BTs than demersal species, Furthermore, purse eye scad and double spotted queenfish from Parit Jawa where is the area of heavy ship traffic line at the Strait of Malacca showed the highest BTs concentrations among fish. The average percentage of TBT in total BTs in fish was 66% and the ratio of each BT was higher in the order of TBT>MBT>DBT, being similar to BTs composition in mussels.

Risk for humam

The biological samples used in the present study are important source for diet. Therefore, the health risk for human by BT in biological samples is of concern. Penninks (1993) derived a tolerable daily intake (TDI) of TBTO of 0.25 µg/kg body weight/day. This value based on the observed effects of

Table 4. The concentration of organotin compounds in fish, 1998/μg Sn kg^{-1} (Sudaryanto et al., 2004) All values are converted to Sn ions.

Locations	Fish	MBT	DBT	TBT
Kotabahru	Mackerel tuna	3.6	6.3	71
	Finny scad	2.6	1.8	4.2
	Rosy threadfin bream	3.9	1.6	2.4
Langkawi	Finny scad	3.9	2.6	8.3
	Purse eye scad	3.9	2	15
	Patterned tongue sole	2.5	<1.3	2.8
	Redatil scad	2.3	2.3	10
Kuala Terengganu	Purse eye scad	3.4	2.1	13
	Redatil scad	5.8	1.9	8.7
Mersing	Spotted javelinfish	4.2	1.8	3.8
	Long-jawed mackerel	4.2	6.2	69
	Purse eye scad	4.9	4.8	15
Parit Jawa	Purse eye scad	5.7	13	72
	Double spotted queenfish	6.3	11	190
Port Dickson	Black pomfret	6.3	2.9	36
	Finny scad	7.4	11	59
	Purse eye scad	3.9	2.5	30

TBT on the immune function in rats with a safety factor of 100 to extrapolated the toxicity test from rats to humans. Tolerable average residue levels (TARL) was calculated from TDI based the formula as described below.

TARL=(TDI×60 kg body weight)/average daily seafood consumption) ············ (1)

Tolerable average residue levels of TBT for seafood in Malaysia were estimated to be 40.9 μg kg^{-1} wet weight for an average person weighing of 60 kg and daily seafood consumption of 146.6 g (Belfroid et al. 2000). Acceptable daily intake (ADI) was proposed to be 0.5 μg/kg/body weight by World Health Organization (FAO/WHO, 1971). Acceptable concentration of TPT was calculated from formula (1) using ADI instead of TDI. Acceptable concentration of TPT was 63 μg kg^{-1}. The concentrations of TBT in some fish samples also exceeded TARL. TPTs were not detected in biological samples.

The current status of imposex

Tan (1997) reported incidence of imposex of *T. clavigera* in Singapore site of the Strait of Johor in 1993. The relative penis size (RPS) index was 18.8% and proportion of females with aborted egg capsules was 4%. Swennen et al. (1997) collected the gastropods in the Strait of Malacca between Kuala Selangor and Singapore and surveyed the proportion of imposex in gastropods. The highest incidence was found in the Southern part of the Strait of Malacca and imposex could be found up to nearly 30 miles from the shipping route in the Strait of Malacca. However, no relation was found between imposex incidence and sampling deph (0–30 m). These reports show that Malaysia is considerable contaminated by TBT.

Conclusion

Judging from the concentration of OTs in aquatic environment and incidence of imposex, the contamination by OTs is more serious issues in Malaysia. The concentrations of OTs in water and mussels will be decreasing in near future by the worldwide ban of OTs. On the other hand, OTs in sediment will persist for long time, because TBT is not degraded in sediment (Maguire et al. 1983, Dowson et al. 1993). Therefore, the OTs contamination in the area with heavy traffic of ships such as the Strait of Malacca and the Strait of Johor will not recover after the ban of OTs. The dredging of OTs contaminated sediment will be needed in these areas, in order to remediate the Malaysian environment from OTs contamination.

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