Review:

A REVIEW OF ANTIFOULING BIOCIDES CONTAMINATIONS IN INDONESIA, MALAYSIA, THAILAND AND VIETNAM

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ABSTRACT

The concentrations and the distribution of organotin (OT) compounds and booster biocides in sediment and biological samples from Indonesia, Malaysia, Thailand and Vietnam are reviewed. Basically, the concentrations of tributyltin (TBT) in these countries were within the levels that have been reported in developed countries. However, the concentrations of triphenyltin (TPT) and booster biocides were lower than the reported values. Concerning the spatial distribution of antifouling biocides, higher concentrations of OTs were detected in the industrial area and fishing ports. Among ASEAN countries, the highest concentrations of TBT and TPT were observed in Bitung, Indonesia, where many ships from Southeast Asia visit and is moored in the port. The ratio of detected alternative biocide differed from country to country. Especially, many booster biocides were detected and their concentrations were high in Vietnam.

Keywords: sediment, mussel, butyltin, phenyltin, alternative biocides.

INTRODUCTION

Organotin (OT) compounds leaching from antifouling paints have caused many deleterious effects on nontarget aquatic organisms, including imposex and abnormal shell morphology (Gibbs et al., 1988;Waldock and Thain, 1983). As a result, OTs have led to a decrease in aquatic products (Bryan et al., 1986). OT application to large vessels has been banned or restricted in some countries. Even after the enforcement of the regulation, OTs have nonetheless been detected at higher concentrations in water, sediment, and biota from harbours, marinas, and estuaries, particularly where boat activity is high and water is poor flushing (e.g., Harino et al., 1998ab). In October 2001, the International Maritime Organization (IMO) adopted the International Convention on the Control of Harmful Antifouling Systems (AFS Convention), which prohibited the use of OTs as active ingredients in antifouling systems for ships. Following the international restrictions on the use of OT-based antifoulants, paint manufacturers have developed many products as alternatives to the use of OTs. More than 20 chemical substances have been used or proposed as alternative compounds. When these antifouling biocides from the hulls of ships, fishing nets etc. are released into the aquatic

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environment, these chemicals are distributed to water, sediment, and aquatic organisms. In fact, aquatic pollution by these booster biocides has been already reported in some countries and Sea-Nine 211, Diuron, and Irgarol 1051, which were representative booster biocides, were detected at the levels of sub μg l^{-1} and sub μg kg^{-1} dry weight (dw) in water and sediment from the coastal area, respectively (e.g., Scarlett et al., 1999).

Recently, in some ASEAN countries, the economic growth accompanying rapid development of industries has caused an expansion of international trade. This factor increases the potential risk for antifouling biocide contaminations to occur.

The authors and colleagues have investigated the distribution of antifouling biocides in sediment and biological samples from Indonesia, Malaysia, Thailand and Vietnam which are shown in Fig. 1, in "Core University Program of the Japan Society for the Promotion of Science (JSPS), "Coastal Marine Science (CMS)" followed by the Asian CORE program "Establishment of Research and Education Network on Coastal Marine Science in Southeast Asia (COMSEA)" and have got many findings of current status of antifouling biocides in ASEAN countries (Harino et al., 2006ab; 2010; 2012; Midorikawa et al., 2004). In this paper, these findings are reviewed.

INDONESIA

Sediment core samples were collected in Bitung, Gangga Island, Manado and Jakarta Bay in Indonesia in 2004 (Fig. 2). Bitung, Gangga Island and Manado are located in the North Sulawesi countryside. Because the main industry of Bitung is fishery, there are many fishing boats in the bay, and big ships from Malaysia and Singapore frequent and are moored in the port of Bitung. Gangga Island is a resort area that has a white sand beach surrounded by a garden of tropical trees and plants at the base of a jungle-covered-hill. Manado is the capital city of the North Sulawesi province of Indonesia, situated on the Bay of Manado and surrounded by a mountainous area. The main industries are tourism and fishery. Sediment samples were also taken at 20 sampling sites (water depth; J1-J10 2-5m, J11-J20 12-24m) in Jakarta Bay in 2007. Jakarta is the capital and largest city of Indonesia. It also has a greater population than that of any other city in Southeast Asia. Jakarta Bay, at an inlet of the Java Sea, is an important

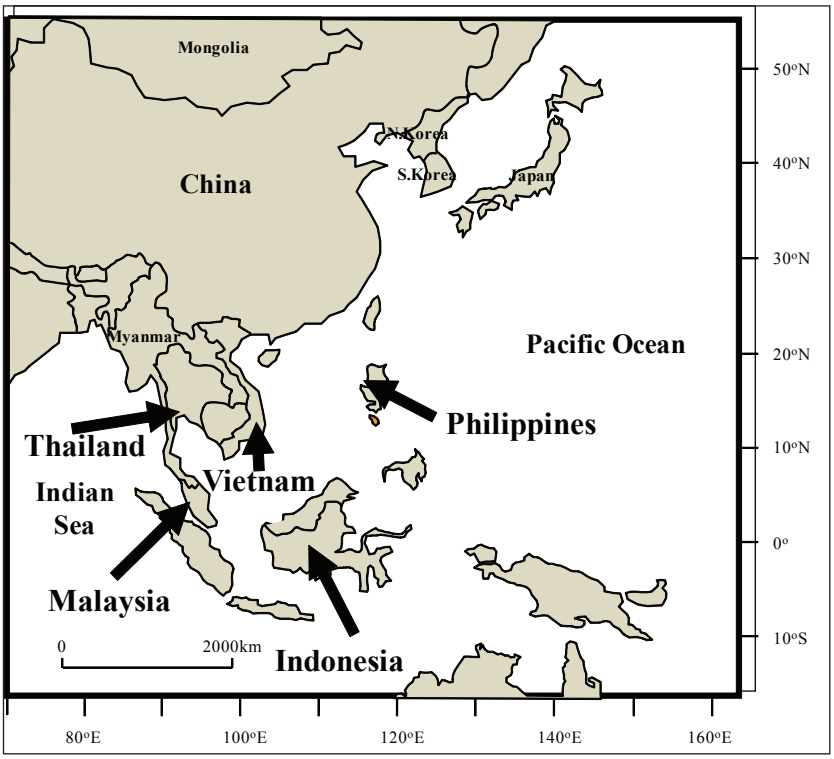

Figure 1. Sampling sites in ASEAN countries.

port for Indonesia. Many small boats are moored in the coastal waters of Jakarta.

Antifouling biocide concentrations are shown in Table 1. Tributyltin (TBT) compounds in subsurface sediment from Indonesian coastal water were detected in the range of 0.4 – 350 μg kg^{-1} dw (means 87μg kg^{-1} dw). The highest concentration of TBT was observed in Bitung. A higher concentration of TBT was also observed in Manado, where the main industry is fishery. TBT was the dominant species among BTs in Bitung and Manado. It is considered that the inputs of TBT in these sampling areas continued. On the other hand, TBT concentrations in Jakarta Bay were low in the range of 0.4 – 52 μg kg^{-1} dw, suggesting that Manado and Bitung are more contaminated by TBT than Jakarta. TBT concentrations in Gangga Island which is a tourist resort area, was the lowest among the sampling sites.

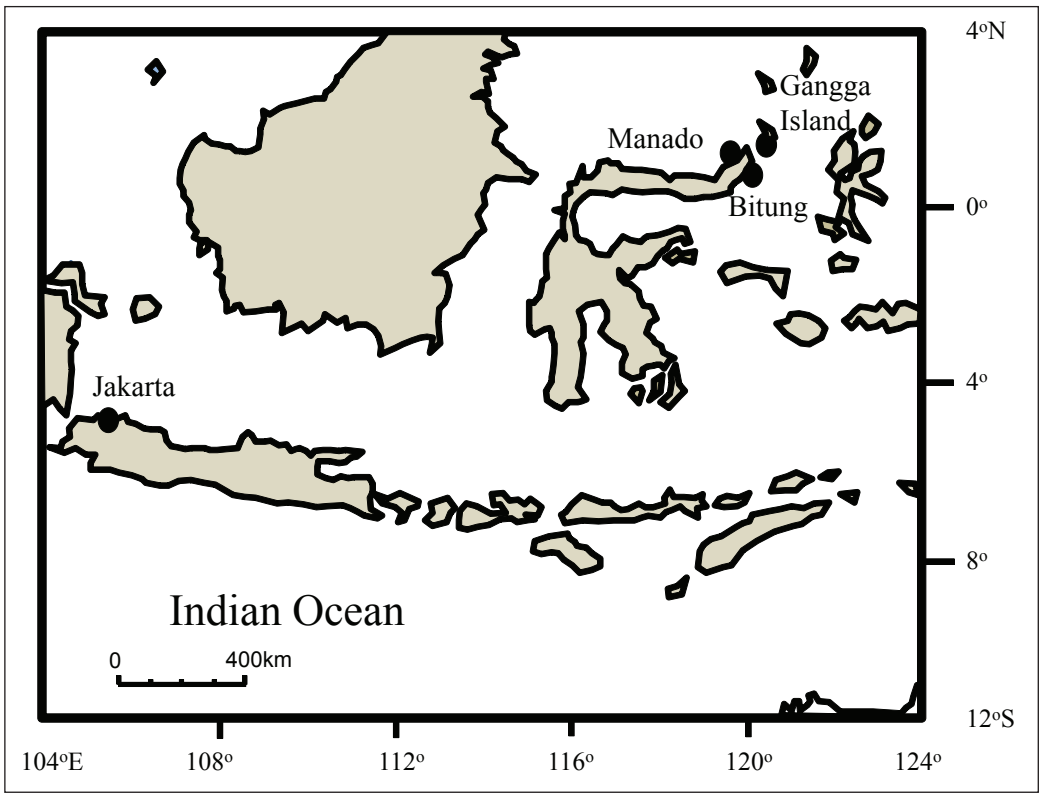

Figure 2. Sampling sites in Indonesia (Source: Harino et al., 2012).

Table 1. The concentrations of alternative biocides in sediment from Indonesia (μg kg^{-1} dw) (Souce: Harino et al., 2012)

	MBT	DBT	TBT	MPT	DPT	TPT	Sea Nine 211	Diuron	Dichlofluanid	Irgarol 1051	M1
Bitung	63 (49-76)	2 (2-3)	250 (160-350)	5 (4-6)	<0.1 <0.1	10 (0-19)	79 (53-110)	574 (410-740)	<0.4 <0.4	63 (45-80)	68 (61-76)
Manado	16	1.9	89	4.5	1.0	<0.1	56	410	<0.4	57	100
Gangga island	1.5	2.2	0.7	3.7	2.4	<0.1	150	210	<0.4	37	670
Jakarta Bay	28 (4.5-170)	12 (0.9-78)	10 (0.4-52)	7.1 (0.2-22)	2.8 (0.1-39)	0.5 (0-7.1)	<0.04 <0.04	<0.04 <0.04	<0.4 <0.4	0.4 (0.1-1.1)	0.4 0.4

Triphenyltin (TPT) compounds in Bitung and Jakarta Bay were detected in the range of <0.1 - 19 μg kg^{-1} and <0.1 – 7.1 μg kg^{-1}, respectively (Table 1). Kannan and Lee (1996) reported that TPT used as a pesticide was detected in contaminated sediment. TPT may be in used currently as a pesticide in Bitung and Jakarta Bay.

The concentrations of Sea Nine 211, Diuron, Irgarol 1051 in sediment from Indonesia were in the range of <0.04 – 110μg kg^{-1} dw, <0.04 – 740 μg kg^{-1} dw and 0.1 – 80 μg kg^{-1} dw, respectively (Table 1). The concentrations of alternative biocides from Indonesia were compared with the reported values. No Sea Nine 211 was detected in sediment from Southampton, England (Thomas *et al,*. 2000). While, Sea Nine 211 was detected at every sampling sites in Japan (Harino et al., 2005; 2007; 2010; Eguchi et al., 2010). The concentrations of Sea Nine 211 in sediment from Japan (the Port of Osaka, Maizuru Bay, Otsuchi Bay and Tanabe Bay) were in the range of <0.04 - 110 μg kg^{-1} dw. The concentrations of Sea Nine 211 in sediment from Indonesia were higher than the values for other countries. The concentrations of Diuron and Irgarol 1051 in sediment have also been reported in many countries The concentrations of Diuron in the sediment from the coastal waters of England were in the range of <12 – 395 μg kg^{-1} dw as reported by Boxall et al. (2000). Thomas et al. (2000; 2002) reported that the Diuron concentrations were in the range of <0.1 – 1.4 μg kg^{-1} dw in Southampton. Harino et al. (2004) reported that Diuron was detected in the range of 0.64 - 1350 μg kg^{-1} dw in the Port of Osaka, Japan. The concentrations of Diuron were greatly different according to sampling sites. The concentrations of Diuron in sediment from Indonesia were within these reported values. The concentrations of Irgarol 1051 in sediment from England, Germany, Switzerland, Japan, were in the ranges of 6.3 – 880 μg kg^{-1} dw, 12.5 – 67.8 μg kg^{-1} dw, <0.2 – 8 μg kg^{-1} dw and <0.05 – 21μg kg^{-1} dw, respectively. (Harino et al., 2005, 2006a, b; 2007; Boxall et al., 2000; Gough et al., 1994; Thomas et al., 2002; Voulvoulis et al., 2000; Biselli et al., 2000; Toth et al., 1996). Thus, the Irgarol 1051 concentrations in the sediment from Indonesia were within the range reported for coastal waters in other countries.

Diuron concentrations in Bitung, Manado and Gangga Island were higher than those of the other alternative biocides and Irgarol 1051 were also detected in Jakarta Bay (Table 2). It is evident that alternative biocides have accumulated in sediment in Indonesia as well as TBT.

Butyltin (BT) compounds concentrations at Sts. J6 and 8, where the small boats are moored, were high (Fig. 3). Although TBT were detected in the coastal area of Jakarta Bay, monobutyltin

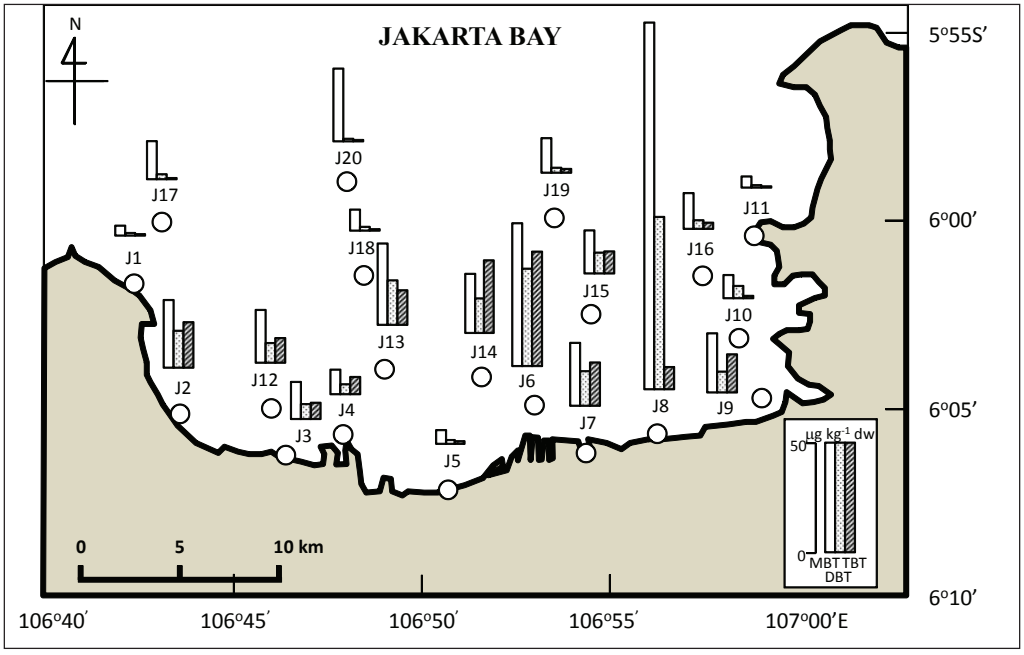

Figure 3. Distribution of BTs in Jakarta Bay (Source: Harino et al., 2012).

(MBT) was the dominant species among BTs at all of the sampling sites, suggesting that TBT is not used now and TBT degraded in sediment (Fig. 3). The concentrations of phenyltin (PT) compounds were lower than those of BTs, and the degradation products of TPT were dominant rather than TPT (Fig. 4). The concentrations of PTs were high at Sts J2-3, which are near mangrove forests. It was considered that PTs used as a pesticide have been discharged to the coastal area (Kannan and Lee, 1996).

Although the concentrations of Irgarol 1051 from Jakarta Bay were in the range of <0.04-1.1 μg kg^{-1} dw as shown in Table 2, the other alternative biocides were not detected. Irgarol concentrations were high at St. J8 where TBT concentrations were high. This indicates that Irgarol 1051 is used as a substitute for TBT.

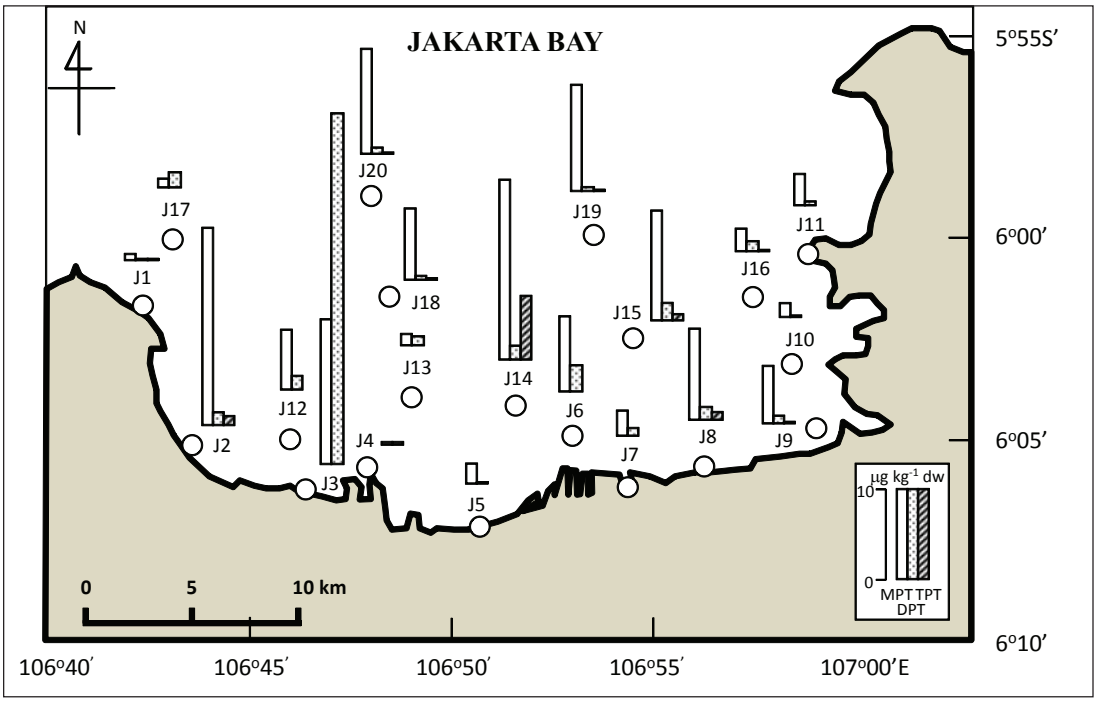

Figure 4. Distribution of PTs in Jakarta Bay (Source: Harino et al., 2012).

Table 2. Concentrations of antifouling biocides in sediment from Peninsular Malaysia/μg kg^{-1} dw (Source: Harino et al., 2009)

Location	MBT	DBT	TBT	MPT	DPT	TPT	Seanine-211	Diuron	Dichlofluanid	Irgarol 1051	M1	Pyrithions
P1	19	7.1	19	16	0.4	0.2	0.05	<0.02	<0.1	<0.02	<0.09	<20
P2	17	7.9	8.9	77	24	28	<0.04	0.39	<0.1	1.2	<0.09	<20
P3	19	8.8	15	32	10	13	<0.04	0.12	<0.1	0.25	<0.09	<20
P4	24	9.7	20	<0.1	1.2	0.8	<0.04	0.29	<0.1	<0.02	<0.09	<20
P5	5.5	1.4	0.9	30	8.2	11	<0.04	0.08	<0.1	0.2	<0.09	<20
P6	14	4.7	1.9	81	19	24	<0.04	0.14	<0.1	0.40	<0.09	<20
P7	5.3	1.3	1.5	<0.1	0.4	0.1	<0.04	<0.02	<0.1	0.17	<0.09	<20
P8	4.1	1.1	0.7	<0.1	0.5	0.2	<0.04	<0.02	<0.1	<0.02	<0.09	<20
P9	12	2.5	1.1	44	17	21	<0.04	<0.02	<0.1	0.80	<0.09	<20
P10	42	13	160	65	4.2	0.8	1.7	4.8	<0.1	14	<0.09	<20
P11	25	20	150	<0.1	0.7	0.4	<0.04	<0.02	<0.1	0.25	<0.09	<20
P12	46	23	41	121	27	34	<0.04	0.86	<0.1	1.9	<0.09	<20
P13	240	190	230	<0.1	1.7	0.9	<0.04	4.3	<0.1	2.6	<0.09	<20

MALAYSIA

Malaysia is located in the center of Southeast Asia, and consists of two geographical regions, the Peninsular Malaysia and Malaysian Borneo divided by the South China Sea. Mining has declined but the electronic industry has flourished, and as result, Malaysia has become famous in Southeast Asia as an exporter of electronic parts. In order to confirm the widespread OTs contaminations, sediment and biological samples were taken in Peninsular Malaysia along the Strait of Malacca from September 7th to 15th, 2006 (Fig. 5). The surface sediment samples were taken using a core sampler in 13 sites. The green mussel (*Perna viridis*) samples were taken at 10 sites. The shell lengths of the green mussels were in the range of 61 – 110 mm. Sediment samples were also taken in the coastal area of Melaka and the Strait of Johor from September 28th to 30th, 2005 (Fig. 5).

The coastal area of Peninsular Malaysia, which is called the commercial capital is an area where many ships sail and are moored. Figure 6 shows the spatial distribution of BTs in the sediment from Peninsular Malaysia along the Strait of Malacca. The concentrations of BTs were high in the southern part of Peninsular Malaysia. In the northern part of Peninsular Malaysia, tourism is a big industry because there are many mangrove forests and beautiful beaches, while in the southern part, there are fishing grounds, fish farms and industrial ports. Severe BTs contamination was therefore detected in the southern part. The concentrations of MBT, dibutyltin (DBT) and TBT in sediment from the coastal waters of Peninsular Malaysia along the Strait of Malacca were in the range of 4.1–242 μg kg^{-1} dw, 1.1 -186 μg kg^{-1} dw and 0.7–228 μg kg^{-1} dw, respectively (Table 2). The maximum concentration of TBT was observed at St. P13, where there is heavy ship traffic. The TBT concentrations in sediment from Sts. P10-11, which contains aquaculture and fishery areas, were also high. We predicted that a lot of TBT

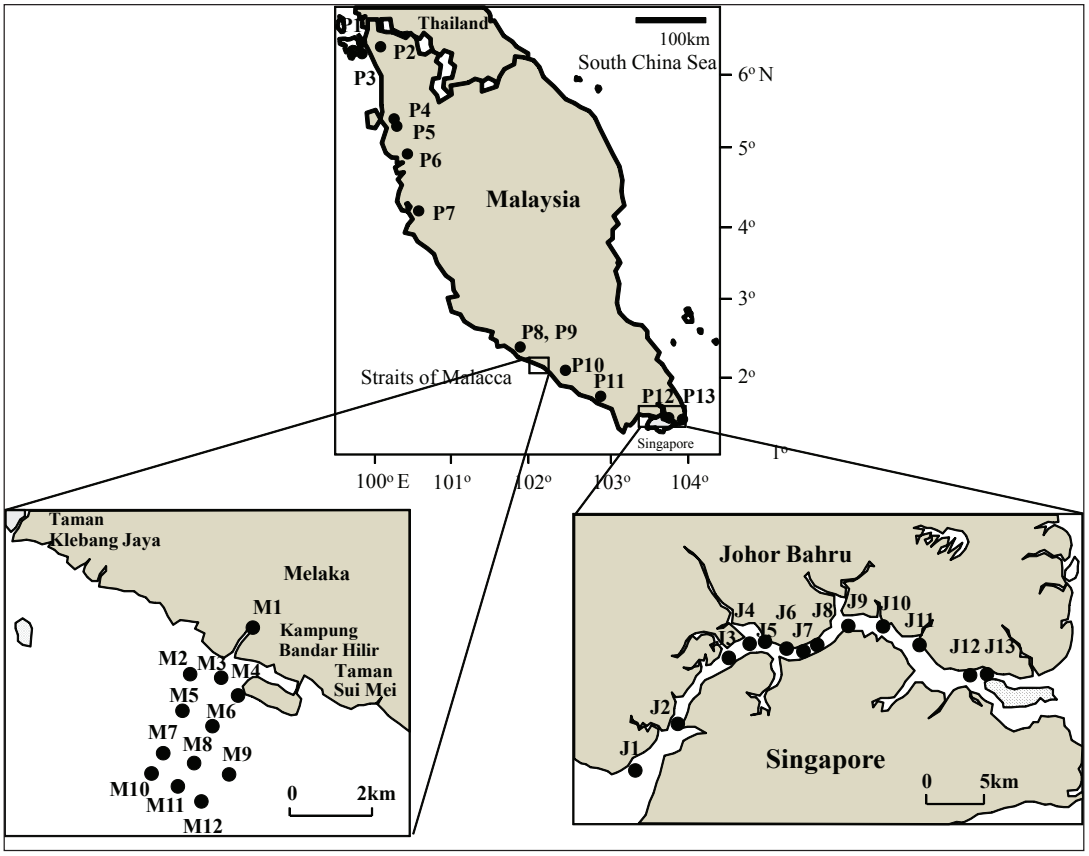

Figure 5. Sampling sites in Malaysia (Source: Harino et al., 2009).

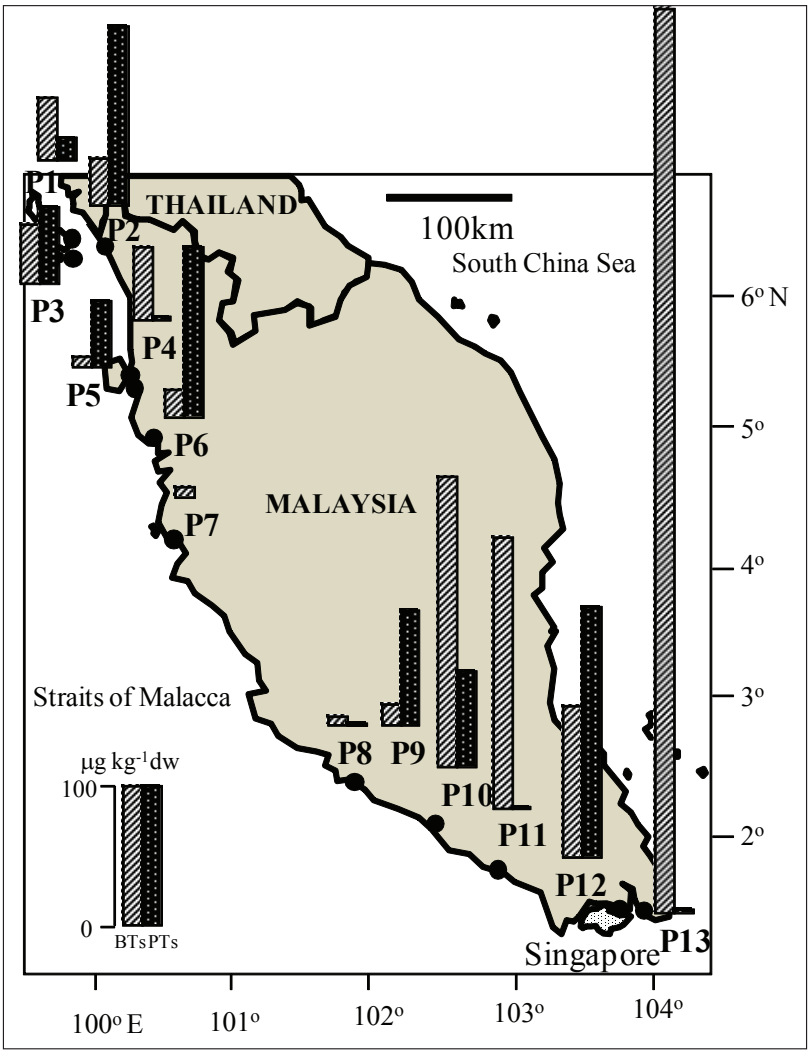

Figure 6. The spatial distribution of BTs and PTs in sediment from Peninsular Malaysia (Source: Harino et al., 2009).

was used in this area in the past. Furthermore, the high percentages of TBT in sediment from Sts. P10–11 and P13 suggest that TBT is still being used in these areas.

The spatial distribution of PTs in Peninsular Malaysia is shown in Fig. 6. Despite the detection of PTs in most sites, a characteristic trend was not observed in the spatial distribution. The concentrations of monophenyltin (MPT), diphenyltin (DPT) and TPT were in the range of <0.1–121 μg kg^{-1} dw, 0.4–27 μg kg^{-1} dw and 0.1–34 μg kg^{-1} dw in the sediment from the coastal waters of Peninsular Malaysia, respectively. Of total PTs, MPT was the dominant species. Kannan and Lee (1996) reported that TPT was used as a pesticide. The detections of PTs in Peninsular Malaysia may be due to its use as a pesticide and not as an antifouling biocide, because the spatial distribution of PTs is different from that of BTs.

Fig. 7 shows the spatial distribution of BTs and PTs in green mussels from Peninsular Malaysia. Differences in BTs levels among sites were not observed. It was reported that TBTs in mussels reflected on those in water within 2 or 3 months (Short and Sharp, 1989). It is presumed that BTs in water are at similar levels among the sampling sites. MBT, DBT and TBT in green mussel samples were detected in the range of 41–102 μg kg^{-1}, 3–5 μg kg^{-1} and 8–32 μg kg^{-1}, respectively. Of all BTs, the percentage of TBT was low, suggesting the decrease of TBT input. Although MPT and DPT in mussel samples were detected in the range of <0.1–22 μg kg^{-1} and <0.1 -5 μg kg^{-1}, respectively, TPT was not detected.

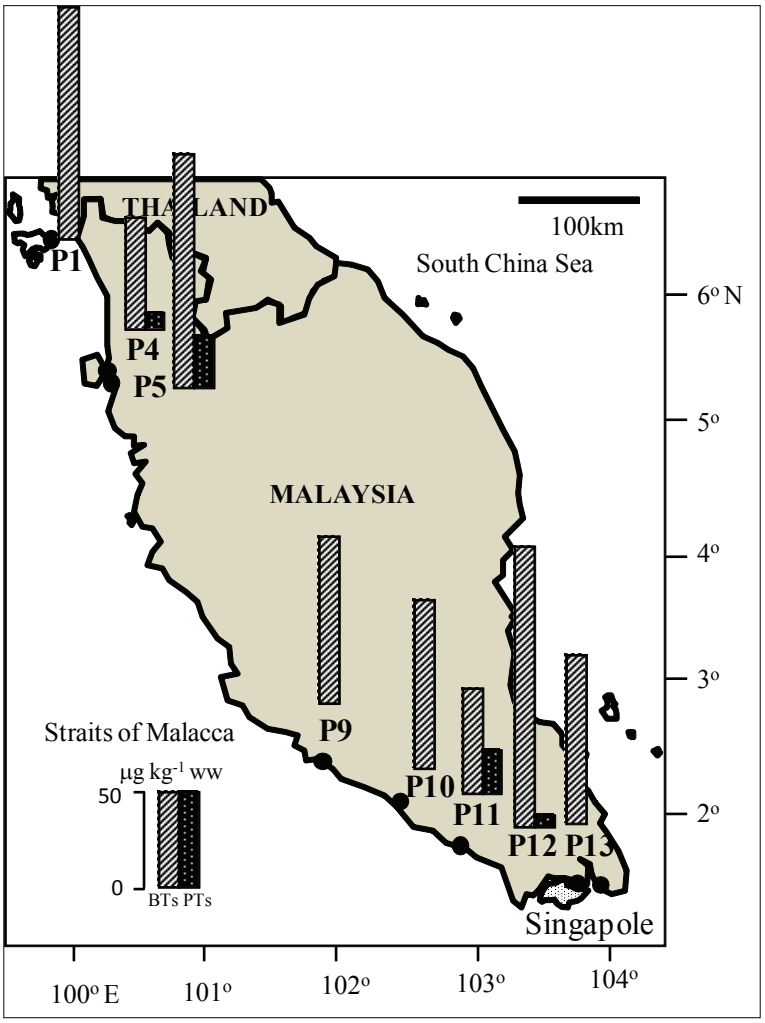

Figure 7. The spatial distribution of BTs and PTs in mussel from Peninsular Malaysia (Source: Harino et al., 2009).

The mussels used in the present study are an important source of food. The tolerable average residue levels (TARL) was calculated using the values of a tolerable daily intake (TDI) of TBTO of 0.25 μg/kg body weight/day (Penninks,1993), an average person weighing of 60 kg and consuming of 146.6 g of seafood a day (Belfroid et al., 2000). TARL for seafood in Malaysia were estimated to be 20.4 μg kg^{-1} ww. The maximum value of TBT which was detected in mussel samples was higher than TARL, suggesting the importance of further monitoring of the TBT concentrations in mussel samples.

As alternative compounds, Sea Nine 211, Diuron, Irgarol 1051, M1 and Pyrithione were measured in sediment from Peninsular Malaysia. The concentrations of Diuron in the sediment were in the range <0.1–5 μg kg^{-1} dw. The concentrations of Diuron were lower than the reported values described in the previous section (Boxall et al., 2000; Thomas et al., 2000, 2002; Harino et al., 2004). The concentrations of Irgarol 1051 in sediment from Malaysia were in the range of <0.1–14 μg kg^{-1} dw. The concentrations of Irgarol 1051 have been reported in various countries. The concentrations of Irgarol 1051 in Malaysia were lower than the reported values in the previous section (Harino et al., 2005; 2006a; 2006b; 2007; Boxall et al., 2000; Gough et al., 1994; Thomas et al., 2002; Voulvoulis et al., 2000; Biselli et al., 2000; Toth *et al,*. 1996). Higher concentrations of Diuron and Irgarol 1051 were observed in Sts. P10, P12-13 where the concentrations of TBT were high. This means that Diuron and Irgarol

1051 are used as an antifouling paint in Malaysia. Sea Nine 211 and Dichlofluanid in sediment from most sampling sites were not detected. However, the concentrations of alternative compounds were lower than those of TBT (Table 3). Namely, the contamination by OTs in Malaysia is a more serious problem than that by alternative compounds.

The concentrations of antifouling biocides in Melaka, where there is great traffic density, were investigated in detail. The concentrations of BTs in sampling sites of Melaka are shown in Table 3. The concentrations of MBT, DBT and TBT in sediment from Melaka were in the range of 85–270 μg kg^{-1} dw, 3.6–17 μg kg^{-1} dw and 2.4–31 μg kg^{-1} dw, respectively, which were similar to those in Peninsular Malaysia. MBT was the dominant species in sediment from Melaka. It is well known that TBT persists for a long time in sediment (Maguire and Tkacz, 1985; Dowson et al., 1993; Harino et al., 1998b). However, the higher percentage of MBT among BTs shows the degradation of TBT. It was reported that in most of the locations tested in Malaysia, MBT generally occurred in the highest proportion (Sudaryanto et al., 2004). The degradation of TBT in sediment from Malaysia may be faster than those in other areas because of higher temperatures. BTs were similar values among all sampling sites from Melaka, indicating that BTs contamination spread off coast. The concentrations of PTs were lower than those of BTs and the spatial distribution of PTs in Melaka was different from those of BTs (Table 3). The input of PTs to sediment may be due to its use as a pesticide, as described in the previous section.

Sea Nine 211, Diuron and Irgarol 1051 in the sediment from Melaka were detected in the ranges of <0.04–4.2 μg kg^{-1} dw, <0.02–4.1 μg kg^{-1} dw and <0.02–0.21 μg kg^{-1} dw, respectively, while Dichlofluanid, M1 and Pyrithiones were not detected. The concentrations of Sea Nine 211, Diuron and Irgarol 1051 were high at St. M1, which is located at the mouth of a river. In fact, the spatial distributions of alternative biocides were different from those of BTs. BTs pollution in sediment was spread across a wide area, because BTs have been used for a long time. However, the contamination by alternative compounds showed localized distribution, because alternative biocides have recently begun to be used.

The Strait of Johor is also an area with great traffic density. The spatial distribution of BTs in sediment from the Strait of Johor is shown in Fig. 8 and Table 4. BTs concentrations in sediment were high at St. J7–J11 with poor flushing of water. The concentrations of MBT, DBT and TBT in the Strait of Johor were in the ranges of 83–542 μg kg^{-1} dw, 30–232 μg kg^{-1} dw and 41–492 μg kg^{-1}

Table 3. Concentrations of antifouling biocides in sediment from Melaka/μg kg^{-1} dw (Source: Harino et al., 2009).

Location	MBT	DBT	TBT	MPT	DPT	TPT	Seanine-211	Diuron	Dichlofluanid	Irgarol 1051	M1	Pyrithions
M1	200	17	31	9.2	0.5	<0.1	4.2	4.1	<0.1	0.21	<0.09	<20
M2	140	5.2	4.1	5.9	0.4	1.3	0.11	0.18	<0.1	0.15	<0.09	<20
M3	120	5.9	8.1	10	0.3	1.1	0.11	<0.02	<0.1	0.05	<0.09	<20
M4	85	3.6	2.4	4.8	0.2	0.4	0.11	<0.02	<0.1	0.06	<0.09	<20
M5	120	5.7	3.8	3.6	0.3	0.3	<0.04	<0.02	<0.1	0.08	<0.09	<20
M6	260	7.5	4.7	5.6	0.5	1.3	0.14	<0.02	<0.1	0.06	<0.09	<20
M7	150	5.3	4.1	3.7	0.3	1.0	<0.04	0.48	<0.1	0.06	<0.09	<20
M8	270	7.0	4.3	11	0.5	1.6	0.09	0.36	<0.1	0.06	<0.09	<20
M9	130	5.8	2.9	8.0	0.1	0.3	0.20	<0.02	<0.1	<0.02	<0.09	<20
M10	170	7.5	4.4	4.6	0.1	0.2	0.04	0.99	<0.1	0.11	<0.09	<20
M11	150	5.6	3.0	6.1	0.1	0.4	<0.04	<0.02	<0.1	0.07	<0.09	<20
M12	230	7.3	8.5	6.4	0.1	0.9	0.09	<0.02	<0.1	0.14	0.49	<20

dw, respectively, which were higher than those in Peninsular Malaysia and Melaka (Table 4). MBT was the dominant compound at most sampling sites of the Strait of Johor, suggesting that the degradation rate of TBT is faster than the input rate in sediment from this area. The concentrations of MPT, DPT and TPT were in the range of 41–66 μg kg^{-1} dw, 5–29 μg kg^{-1} dw and 0.3–34 μg kg^{-1} dw from the Johor Strait, respectively (Fig. 8). The concentrations of PTs in the Strait of Johor were similar to those in the coastal waters of Peninsular Malaysia and Melaka and MPT among PTs was the dominant compound in Johor Strait.

Sea Nine 211, Diuron and Irgarol 1051 in the sediment from the Strait of Johor were detected in the range of <0.04–0.92μg kg^{-1} dw, <0.02–9.9 μg kg^{-1} dw and <0.02–0.90 μg kg^{-1} dw, respectively and Dichlofluanid, M1 and Pyrithiones were not detected. The Diuron concentration was the highest among alternative biocides.

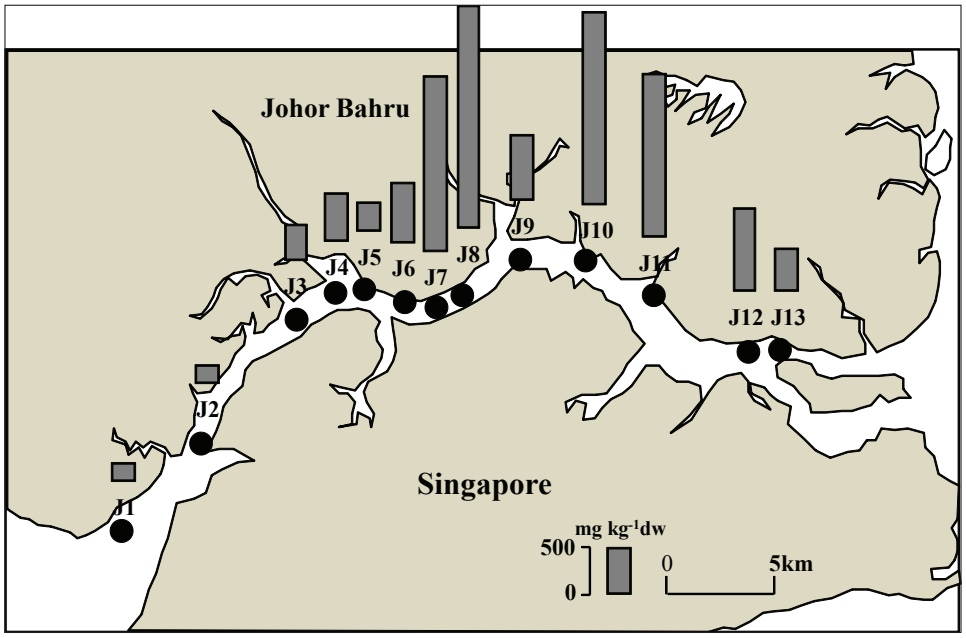

Figure 8. The spatial distribution of BTs in sediment from Johor strait (Source: Harino et al., 2009).

Table 4. Concentrations of antifouling biocides in sediment from The strait of Johor/μg kg^{-1} dw (Source: Harino et al., 2009).

Location	MBT	DBT	TBT	MPT	DPT	TPT	Seanine-211	Diuron	Dichlofluanid	Irgarol 1051	M1	Pyrithions
J1	180	7.1	7.2	3.3	0.2	0.2	0.04	0.22	<0.1	0.04	<0.09	<20
J2	150	19	20	12	0.4	0.8	<0.04	1.0	<0.1	0.11	<0.09	<20
J3	320	26	38	11	0.3	0.7	<0.04	0.89	<0.1	0.11	<0.09	<20
J4	460	32	28	15	0.5	1.1	0.04	0.99	<0.1	0.11	<0.09	<20
J5	290	14	12	15	0.2	0.6	<0.04	1.2	<0.1	0.08	<0.09	<20
J6	480	90	87	8.2	<0.1	<0.1	<0.04	1.8	<0.1	0.13	<0.09	<20
J7	1,300	240	400	10	0.7	<0.1	<0.04	<0.02	<0.1	<0.02	<0.09	<20
J8	1,000	590	880	25	0.7	4.9	0.31	<0.02	<0.1	0.21	<0.09	<20
J9	360	170	200	13	0.6	<0.1	0.15	2.3	<0.1	0.34	<0.09	<20
J10	1100	400	640	38	0.8	2.1	0.92	4.5	<0.1	0.90	<0.09	<20
J11	640	230	940	34	1.7	1.2	0.47	9.9	<0.1	0.75	<0.09	<20
J12	550	110	240	15	0.5	1.3	0.44	3.0	<0.1	0.32	<0.09	<20
J13	350	51	61	4.3	0.2	6.0	0.10	2.1	<0.1	0.14	<0.09	<20

THAILAND

The Kingdom of Thailand lies in the heart of Southeast Asia, making it a natural gateway to Indonesia, Myanmar and Southern China. The coastal line of Thailand located in the southern part of the country has a tropical climate. The survey of antifouling biocides contaminations was conducted along the coastal line of Thailand from April 19th to April 20th, 2004. The sampling sites are shown in Fig. 9. Sediment samples and green mussels (*Perna viridis*) were collected at these sampling sites. The sediment samples were taken using a core sampler in 13 sites. The green mussel samples were taken at 6 sites, except for A1–4, A6, A8 and A13. The shell lengths of the green mussels were in the range of 51–82 mm.

Higher concentrations of TBT in sediment were observed at Sts. A3-4 and A10 (Table 5). In

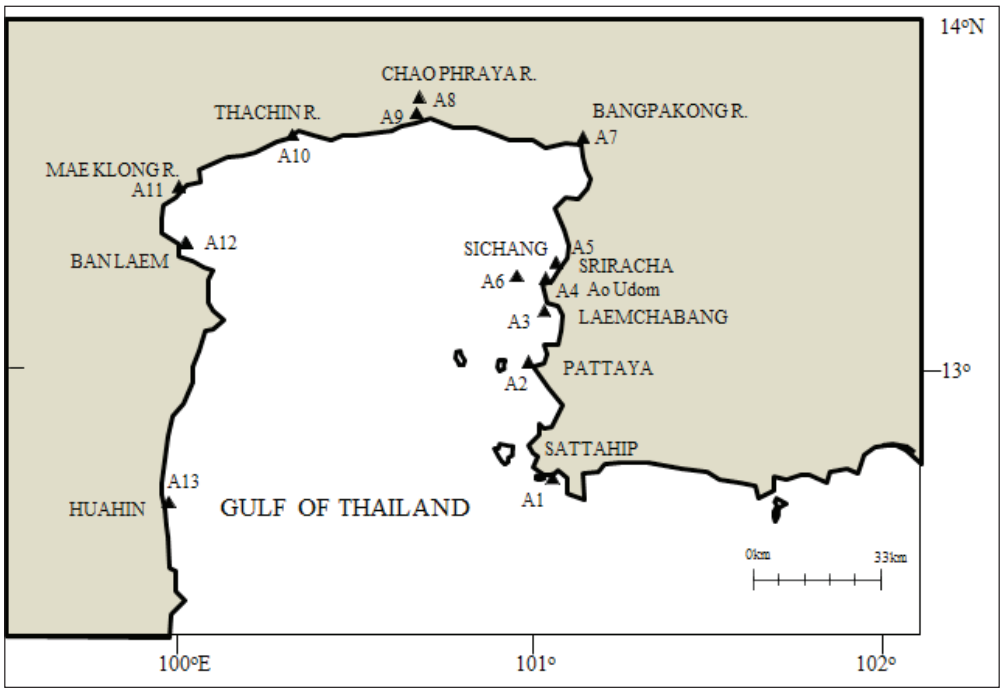

Figure 9. Sampling sites in Thailand (Source: Harino et al., 2006a).

Table 5. Concentrations of antifouling biocides in sediment (μg kg^{-1} dw) from Thailand (Source: Harino et al., 2006a).

		MBT	DBT	TBT	MPT	DPT	TPT	Sea-Nine 211	Diuron	Irgarol 1051	M1
A1	Sattahip, Chonburi	25	38	72	<1	<1	<1	<0.04	2.9	3.2	0.89
A2	Pattaya, Chonburi	10	8	5	<1	<1	<1	<0.04	0.48	0.13	4.9
A3	Laemchabang, Chonburi	59	106	284	<1	<1	<1	<0.04	1.3	0.05	<0.1
A4	Ao Udom, Chonburi	84	85	285	<1	6	29	<0.04	0.12	0.73	<0.1
A5	Sriracha, Chonburi	6	4	3	9	<1	<1	0.09	<0.08	0.09	0.13
A6	Sichang, Chonburi	37	20	34	6	1	12	0.07	0.26	0.07	0.20
A7	Bangpakong River mouth	293	77	28	<1	<1	28	<0.04	3	0.98	0.58
A8	Chao Phraya River mouth	27	19	43	<1	2	<1	<0.04	25	0.85	0.29
A9	Chao Pharaya River mouth	3	6	10	<1	<1	5	<0.04	3.0	0.17	0.20
A10	Thachin River mouth	158	368	1246	<1	<1	7	<0.04	5.7	0.07	0.09
A11	Maeklong River mouth	11	5	10	<1	<1	<1	<0.04	1.5	0.05	0.51
A12	Ban Laem, Petchburi	13	5	11	<1	<1	<1	<0.04	1.5	0.07	0.18
A13	Hua Hin, Prachuabkhirikhan	1	1	2	7	3	7	<0.04	0.20	0.03	0.14

particular, the highest concentration of TBT (1,246 μg kg^{-1} dw) was displayed at station A10, which is an industrial area with shipyards, suggesting the deposit of paint chips containing TBT to sediment. The ratio of TBT among BTs was over 60% at Sts. A3-4 and A10 where TBT concentrations were high. Higher concentration and ratio of TBT in sediment indicate continuous loads of TBT from sailing and mooring ships in stations A3-4 and A10 starting in the past and continuing to the present. The concentrations of PTs were below the detection limit at the most of the stations.

BTs were also detected in green mussels from Thailand (Table 6). The concentrations of TBT ranged from 4 to 45 μg kg^{-1} wet weight (ww). The horizontal distribution of TBT in green mussels will be discussed. The highest concentration in station A5, an aquaculture area with heavy cargo shipping activities, was observed. Although the main industry at Sts. A5, A7 and 12 is aquaculture, TBT concentrations were low. This provides evidence that TBT comes from shipping activities rather than from the use of TBT paint in fishing equipment. The ratios of the degradation products of TBT were higher than those of TBT in green mussels from Sts. A7 and A10–12. Because of a low cytochrome P-450 content and mixed function oxygenase activity in mollusks, mollusks such as green mussels have a limited ability to metabolize TBT (Lee 1986, 1991; Kan-atireklap et al., 1997). Nevertheless, the ratios of MBT were high at most of the sampling sites.

Three species of representative booster biocides (Sea Nine 211, Diuron, and Irgarol 1051) and Irgarol's degradation product's M1 were analyzed in sediment and green mussels from Thailand. Table 6 shows the concentrations of these booster biocides in sediment samples. The detection frequencies of Sea Nine 211 were low (2/13). Furthermore, Sea Nine 211 concentrations in the detected samples were at values near the detection limit (0.051–0.094 μg kg^{-1} dw). The range of Diuron concentrations in sediment from Thailand (0.07–25 μg kg^{-1} dw) was the highest among the booster biocides detected.

Irgarol 1051 was detected in the range of 0.03–3.2 μg kg^{-1} dw M1 of the Irgarol's degradation product was in the range from 0.03 to 4.9 μg kg^{-1} dw. The concentration of Irgarol 1051 in the sediment from Thailand was also the lowest among the reported values (Boxall et al., 2000; Thomas et al., 2000; 2002; Biselli et al.,2000; Harino et al., 2005).

The kinds and levels of detected booster biocides were different at these sample sites. Sea-Nine 211 detected in sediment from Sts. A5-6, which are aquaculture areas and have offshore ports for large cargo vessels. The concentrations of Diuron were over 5 μg kg^{-1} dw at Sts. A8 and A10, which are industrial areas. Irgarol 1051 was found in the highest concentration at station A1, which is a fishing ground and the site of the Royal Thai Navy Base.

Irgarol 1051 is degraded to M1 in the aquatic environment. Therefore, the proportion of M1 for the concentration of Irgarol 1051 was calculated in the sampling sites. The ratios of M1/Irgarol 1051 were greater than one in 8 sites out of the 13 sites. Harino et al. (2005) reported that the concentrations of Irgarol 1051 were generally higher than M1 in sediment from the port of Osaka, Japan. The difference in the pattern between Irgarol 1051 and M1 may be due to the difference of climate

Table 6. Concentrations of antifouling biocides in mussel (μg kg^{-1} ww) from Thailand (Source: Harino et al., 2006a).

		MBT	DBT	TBT	MPT	DPT	TPT	Sea-Nine 211	Diuron	Irgarol 1051	M1
A5	Sriracha, Chonburi	16	9	45	17	52	5	<0.24	<0.64	<0.76	<1
A7	Bangpakong River mouth	13	7	10	10	2	2	<0.24	1.8	<0.76	<1
A9	Chao Pharaya River mouth	10	6	18	31	28	<1	<0.24	9.6	<0.76	<1
A10	Thachin River mouth	8	7	8	25	<1	<1	<0.24	2.5	<0.76	<1
A11	Maeklong River mouth	20	4	4	49	<1	<1	<0.24	1.6	<0.76	<1
A12	Ban Laem, Petchburi	13	4	12	13	<1	1	<0.24	1.2	<0.76	<1

between Thailand, which is tropical, and Japan, which is temperate zone.

The concentrations of booster biocides in green mussels are shown in Table 6. Diuron among booster biocides was only detected with the range of 1.2 – 9.6 μg kg^{-1} ww in sampling sites except for at station A5. The higher water solubility of Diuron (42 mg l^{-1}) means a lower bioaccumulation (United States Environmental Protection Agency, 1989). Nevertheless, Diuron was detected in green mussels at the levels of 1-10 μg kg^{-1} ww, suggesting the relatively higher concentration of Diuron in water.

VIETNAM

Vietnam borders the China to the north, Laos to the northwest and Cambodia to the southwest. With a population of approximately 84 million, Vietnam is one of the most densely populated nations in Southeast Asia. Vietnam has a 3200 km long coastline between 8° 34' N and 21° 33' N, with about 125 beaches of various sizes located along this coastline. The country is composed of northern, central and southern Vietnam. The sediment samples and clams were collected from northern to central Vietnam (Fig. 10). The sediment samples were taken using an Ekman-Birge grab sampler in 9 areas (Tra Co, Cua Luc, Do Son, Ba Lat, Sam Son and Cua Lo in northern Vietnam, and Dong Hoi, Hue and Da Nang in central Vietnam). The clam (*Meretrix* spp.) samples were purchased at local markets in the same sites at which the sediment samples were taken, except for Hue. The shell lengths of the clams ranged from 32–81 mm. The sampling was conducted from March 7–14, 2002 in northern Vietnam and from August 9–14, 2002 in central Vietnam.

Concentrations of MBT, DBT and TBT in sediments from all sampling sites ranged from <0.04 to 11, from 0.64 to 15 and from 0.45 to 34 μg kg^{-1} dw, respectively (Table 7). TBT in the sediment from Hue at 28 μg kg^{-1} dw was highest among north and central Vietnam. Hue is the trading port, which many small vessels are sailed and moored. Furthermore, this sampling site is in a zone with poor flushing. The concentrations of TBT in sediment were high in Cua Luc (15 μg kg^{-1} dw), despite good water flashing, (Table 7). Cua Luc is one of industrial areas with big international trading port. The many vessels including the large hull-vessels from various countries therefore moored and sailed. TBT were also high in Sam Son (6.3 μg kg^{-1} dw) and Tra Co (5.5 μg kg^{-1} dw), which are fishing ports in the zone with poor flushing. It is well known that OT compounds are very stable, concentrations in sediment (Maguire

Table 7. Concentrations of BTs and PTs in sediment from Vietnam (Source: Midorikawa et al., 2004).

Locality	Concentration (μg kg^{-1} dw)					
	MBT	DBT	TBT	MPT	DPT	TPT
Tra Co	2.4	0.79	5.5	2.3	0.65	0.08
Cua Luc	11	4.6	15	5.9	0.29	0.14
Do Son	<0.04	0.78	1.2	3.3	0.16	0.07
Ba Lat	<0.04	0.64	0.89	2.2	0.13	0.06
Sam Son	<0.04	3.2	6.3	5.3	0.22	0.15
Cua Lo	<0.04	1.1	2.2	2.6	0.18	0.08
Dong Hoi	<0.04	1.3 (0.89-1.8)	3.0 (1.2-4.9)	3.2 (2.9-3.5)	0.13 (0.13-0.13)	0.04 (<0.04-0.08)
Hue	<0.04	1.4 (1.3-1.5)	28 (21-34)	2.7 (2.3-3.1)	0.26 (0.25-0.28)	0.36 (0.33-0.39)
Da Nang	<0.04	1.5	3.2	3.6	2.6	0.09

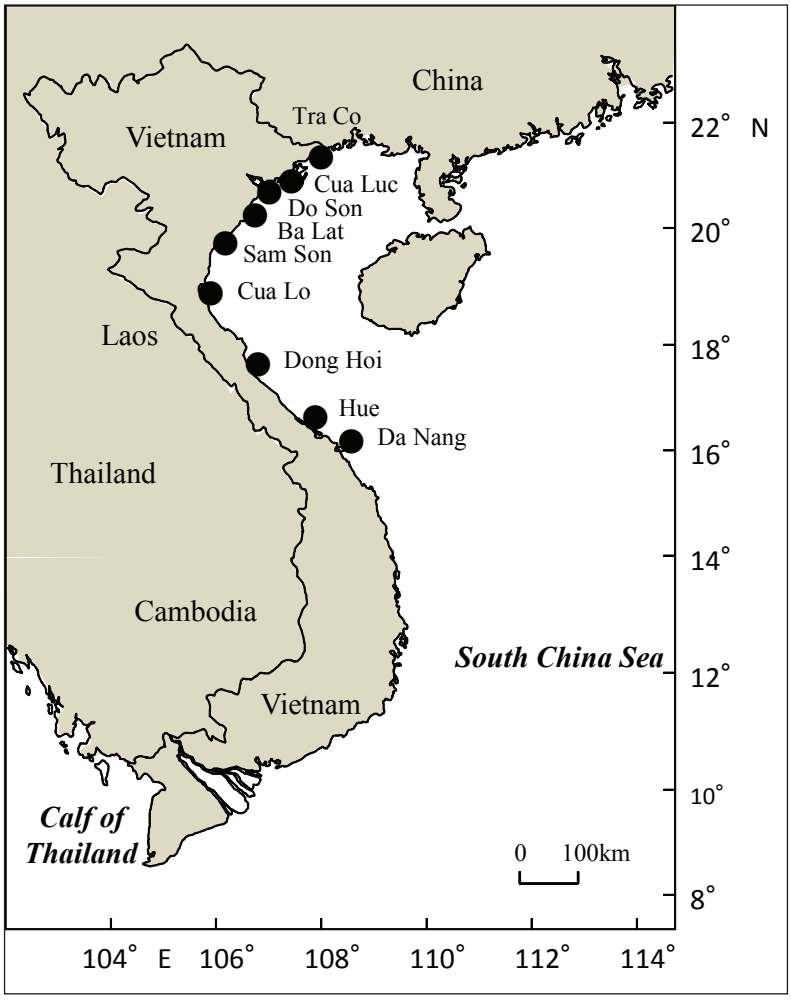

Figure 10. Sampling sites in Vietnam (Source: Midorikawa et al., 2004).

and Tkacz, 1985; Stang et al., 1992). The higher concentration of TBT in sediment from fishing ports and trading ports trading port such as Hue and Cua Luc indicates the continuous input of OTs in these areas. The concentrations of TPT were also high in sediment from Hue (0.36μg g^{-1} dw), followed by Cua Luc (0.14 μg kg^{-1} dw) and Sam Son (0.15 μg kg^{-1} dw) as well as TBT. The higher percentage of TBT among BTs was observed in the all sites, showing continuous input of TBT in these areas.

The levels of PT compounds in sediment from north and central Vietnam were also shown in Table 8. Concentrations of MPT, DPT and TPT in sediments from all sampling sites were in the range of 1.2–5.9, <0.04–3.6 and <0.04–0.39 μg kg^{-1} dw, respectively. These levels of DPT and TPT were close to the detection limit. The concentration of PTs is the order of MPT> DPT>TPT. These results suggest that the degradation rate of TPT in sediment is higher than the accumulation rate of TPT, and TPT input itself is decreasing.

Concentrations of MBT, DBT and TBT in clam from all sites were in the range of 0.1 to 44, 0.5 to10 and 1.4 to 56 μg kg^{-1} ww, respectively (Table 8). The concentrations of TBT from Vietnam were compared to the other countries. The concentrations of TBT in the gaper clams, *Tresus muttallii* and *Mya arenaria* from the Coos Bay estuary, USA were 2.6 and 56.5–100 μg kg^{-1} wet between 1996 and 1997, respectively (Elgethum et al., 2000). TBT in the clam (*Venerupis decussate*) from Spain have been reported to be in the range of 111–450 μg kg^{-1} wet during 1993–1994 (Gomez-Ariza et al., 2000). The concentrations of TBT in the clam (*Potamocorbula amurensis*) from San Francisco Bay, USA were in the range of 123–166 μg kg^{-1} (Pereira et al., 1999). The concentrations

of TBT from Vietnam are within the ranging of the level in clam from the other countries. It was well known that TPT is more stable than TBT in biological samples (Fent and Hunn,1991). Nevertheless, the levels of TBT in clam were also higher than TPT, suggesting more input of TBT to aquatic environment in comparison with TPT. The levels of TBT in the clam from Cua Luc were high dramatically among north and central Vietnam at 47 μg kg^{-1} ww (Table 8). TBT concentrations from the other 7 sites were in the range of 1.4–6.8 μg kg^{-1} ww. TPT in the clam from Cua Luc was also higher than the other sites in clam as well as TBT.

The concentrations of booster biocides in the sediment samples from the coastal areas of northern and central Vietnam were measured (Table 9). The concentrations of Sea-Nine 211, Dichlofluanid, Duiron, Irgarol 1051 and Pyrithiones in sediment were in the range of 0.09–1.3 μg kg^{-1} dry, < 0.10–13 μg kg^{-1} dry, 0.11–3.0 μg kg^{-1} dry, 0.05–4.0 μg kg^{-1} dry and <2–420 μg kg^{-1} dry, respectively. The levels of booster biocides in sediment from northern and central Vietnam were compared to other world-wide sites that have been investigated. The concentrations of Sea-Nine 211 ranged from 0.09 to 1.3 μg kg^{-1} dry in Vietnam. Sea-Nine 211 was detected in the range of < 0.2–2.35 μg kg^{-1} dry in the Port of Osaka, Japan (Harino et al., 2005). The levels of Sea-Nine 211 in Vietnam are within the reported values as described in the section of Indonesia. The concentrations of Diuron in sediment from Vietnam (0.11–3.0 μg kg^{-1} dry) were at a similar level those in the UK, but a lower than level than in Japan. The reported values of Diuron in the world were described the section of Indonesia (Boxall et al., 2000; Thomas et al., 2000; 2002; Harino et al., 2004). Although the maximum concentration of Dichlofluanid in sediment from Vietnam was 13 μg kg^{-1} dry, the detection frequencies were low. Dichlofluanid was detected in the Blackwater estuary of England at < 4.9–688 μg kg^{-1} dry (Voulvoulis et al., 2000). However, Dichlofluanid was not detected in Southampton, England (Thomas et al., 2002). There have been many reports on Irgarol 1051 concentrations (Harino et al., 2005; 2006a,b; 2007; Boxall et al., 2000; Gough et al., 1994; Thomas et al., 2002; Vouvoulis et al., 2000; Biselli et al., 2000; Toth et al., 1996). The concentrations of Irgarol 1051

Table 8. Concentrations of BTs and PTs in clam (*Meretrix* spp) (Source: Midorikawa et al., 2004).

Location	Concentration (μg kg^{-1} ww)					
	MBT	DBT	TBT	MPT	DPT	TPT
Tra Co	3.5 (1.1- 6.1)	1.3 (0.5- 1.7)	6.5 (2.0- 12)	2.5 (0.2- 4.7)	0.11 (<0.1 - 0.3)	0.57 (0.5- 0.6)
Cua Luc	3.2 (2.6- 3.8)	4.1 (3.9- 4.3)	47 (38- 56)	0.7 (0.3- 1.0)	<0.1	2.7 (2.2- 3.1)
Do Son	0.86 (0.1- 1.6)	0.7 (0.7- 0.7)	1.4 (1.4- 1.4)	0.3 (0.3- 0.4)	<0.1	0.6 (0.5- 0.6)
Ba Lat	1.3 (1.2- 1.4)	0.8 (0.7- 0.8)	4.9 (4.7 -5.1)	0.4 (0.3- 0.5)	<0.1	0.5 (0.4- 0.5)
Sam Son	7.4 (0.8- 15)	2.5 (0.5- 5.5)	3.6 (2.0- 4.6)	4.2 (0.2- 7.8)	0.3 (<0.1 - 0.7)	0.5 (0.3- 0.8)
Cua Lo	1.4 (1.2- 1.6)	0.8 (0.8- 0.8)	1.6 (1.5- 1.8)	0.3 (0.2- 0.4)	<0.1	0.7 (0.5- 0.9)
Dong Hoi	29 (21- 44)	3.1 (1.9- 4.8)	2.0 (1.5- 2.2)	6.2 (4.7- 7.3)	0.2 (0.2- 0.3)	0.5 (0.2- 0.7)
Da Nang	28 (5.2- 40)	6.1 (1.6- 10)	6.8 (2.6- 14)	8.4 (3.9- 11)	0.3 (0.2- 0.5)	0.3 (<0.1- 0.4)

Table 9. Concentrations of alternative biocides in Vietnam (Source: Harino et al., 2006b).

	Booster biocides (μg kg^{-1} dw)					
	Sea-Nine211	Dichlofluanid	Diuron	Irgarol 1051	M1	Pyrithiones
Tra Co	0.14	<0.10	1.0	0.27	<0.10	420
Cua Luc	0.76	13	0.11	1.2	<0.10	<2
Do Son	1.3	12	3.0	0.87	<0.10	<2
Ba Lat	0.18	<0.10	0.31	0.05	0.14	<2
Sam Son	0.50	<0.10	1.8	0.44	0.22	<2
Cua Lo	0.24	<0.10	1.3	0.11	0.20	<2
Don Hoi	0.11	<0.10	1.1	4.0	0.43	<2
Hue	1.1	2.6	1.5	0.07	<0.10	<2
Da Nang	0.09	<0.10	1.4	0.83	0.24	<2

in sediment from Vietnam are also lower than the concentrations in other countries. Generally, the levels of booster biocides in Vietnam are low in comparison with those in other countries.

It has been reported that M1 is produced by the degradation of Irgarol 1051 (Liu et al., 1997; Okamura et al., 1999). The concentration of Irgarol 1051 was higher than that of M1 in most of the sampling sites. The same findings were reported by Thomas et al. (2000).

The distributions of booster biocides in northern and central Vietnam are as follows. Higher concentrations of Sea-Nine 211 (1.1–1.3 μg kg^{-1} dry) were observed in Do Son and Hue. Diuron was also high in Do Son. A higher concentration of Dichlofluanid (12-13 μg kg^{-1} dry) was observed in Cua Luc and Do Son. Irgarol 1051 in Don Hoi (4.0 μg kg^{-1} dry) showed the highest concentration among all sampling sites. Pyrithiones was only detected in Tra Co at a concentration of 420 μg kg^{-1} dry. The concentrations of Sea-Nine 211, Dichlofluanid and Diuron were higher in sediment from international trading ports with poor flushing of water, such as Do Son. (Table 10). Higher concentrations of Irgarol 1051 have been observed in fishing ports such as Don Hoi. Liu et al. (1999) reported that Irgarol 1051 was found more frequently in fishery harbours than in marinas. Thus, which booster compounds were detected is likely to depend on the use of the ports. Pyrithiones was detected in Tra Co.

Although the clams from Cua Luc were found to contain Irgarol 1051 at a trace level, about 0.05 μg kg^{-1}, other booster biocides were not detected. The logarithms of the octanol water partitions (Pow) of Sea-Nine 211, Diuron, Dichlofluanid and Irgarol 1051 were 2.8, 2.9, 3.7 and 2.8, respectively (Harino, 2004). These values imply a low degree of bioconcentration. As these clams were for sale in local markets, many people in Vietnam have been eating them. However, the effects of the consumption of the clam on humans may also be limited, because the concentrations of Irgarol 1051 in clams are low.

Comparison of the concentrations of antifouling biocide among Southeast Asian countries

The concentrations of OTs in sediment were compared among ASEAN countries (Figs.11–12). The higher concentrations and the wide distribution of TBT and TPT in sediment from Bitung and Manado in Indonesia, Malaysia and Thailand

show that larger amounts of these compounds have been used in coastal waters from these countries in the past.

The level of each alternative compound in sediment was different in each country (Fig. 13). The higher concentrations were observed in Irgarol 1051 for Malaysia, Diuron for Thailand and Sea Nine 211 for Vietnam. This reflects the fact that the patterns of application of antifouling biocides vary among regions.

Use of TBT for ship hull was banned in the worldwide in 2008. It is predicted that the concentrations of OTs in aquatic environment will decrease and the concentrations of booster biocides will increase. Therefore, further monitoring of antifouling biocides is needed in ASEAN countries, in order to evaluate the effect of AFS convention.

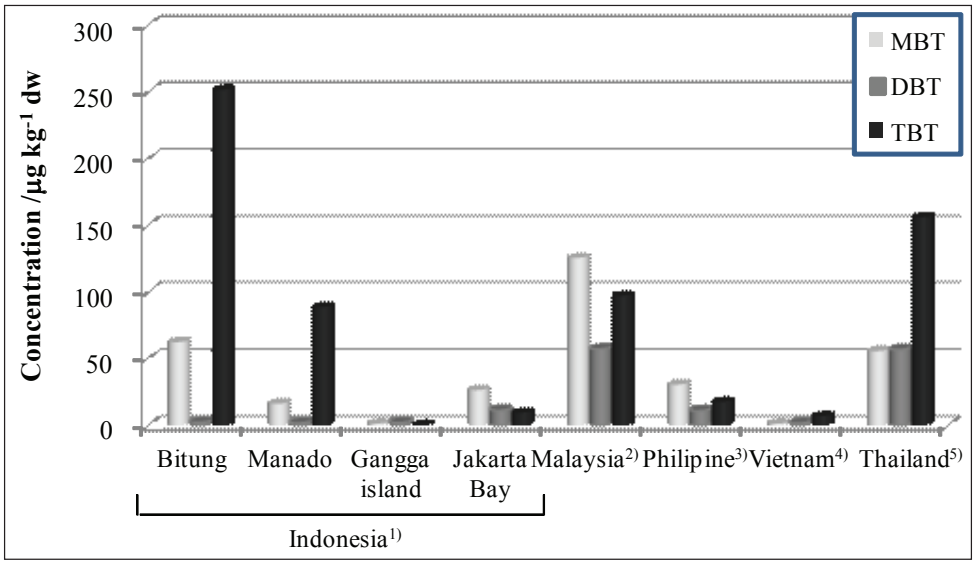

Figure 11. The comparison of BTs concentration in sediment among various countries. 1) Harino et al., 2012, 2) Harino et al., 2009, 3) Harino et al., unpublished data, 4) Midorikawa et al., 2004, 2005) Harino et al., 2006a.

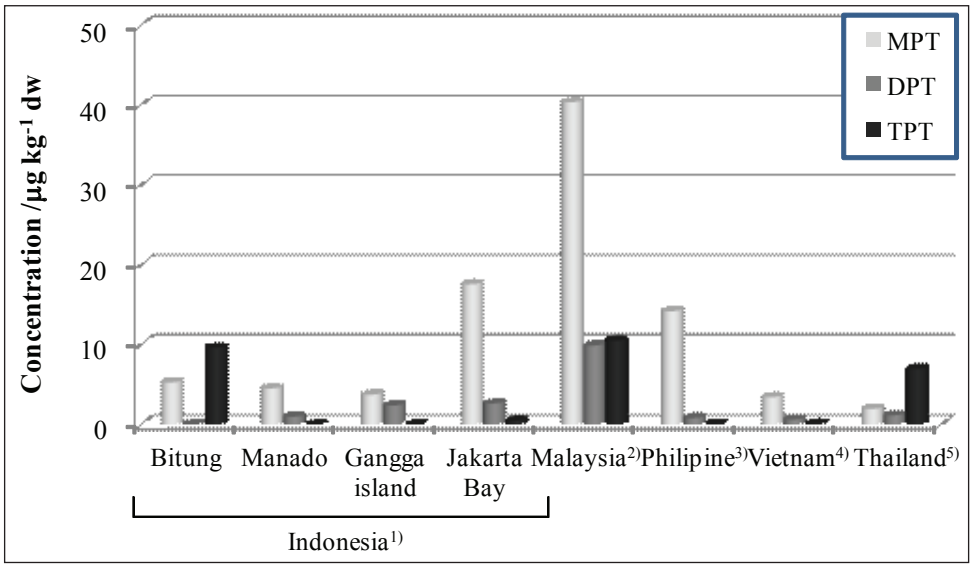

Figure 12. The comparison of PTs concentration in sediment among various countries. 1) Harino et al., 2012, 2) Harino et al., 2009, 3) Harino et al., unpublished data, 4) Midorikawa et al., 2004, 2005) Harino et al., 2006a.

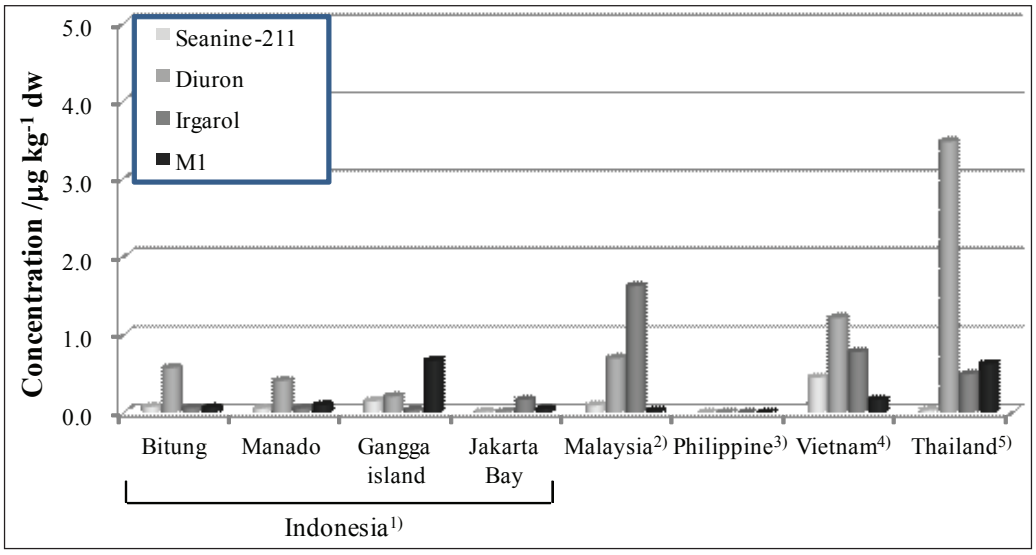

Figure 13. The comparison of antifouling biocides concentrations in sediment among various countries.
1) Harino et al., 2012, 2) Harino et al., 2009, 3) Harino et al., unpublished data, 4) Harino et al., 2006b, 5) Harino et al., 2006a

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