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Status of butyltin contamination in Thailand coastal waters

Gullaya WATTAYAKORN[1,2*]

[1] Aquatic Resources Research Institute, Chulalongkorn University, Bangkok 10330, Thailand.
[2] Department of Marine Science, Faculty of Science, Chulalongkorn University, Bankok 10330, Thailand.
* E-mail: gullaya@chula.ac.th

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Abstract—The aquatic pollution by tributyltin (TBT) arising from anti-fouling paints has been of great concern due to their effect of shell malformation in oysters, mortality of the larvae of mussels and imposex in gastropods, which occur at a very few nanogram per liter of aqueous TBT concentrations. Studies show that TBT is a widespread problem and posing a threat to sensitive species in coastal areas where shipping traffic intensities are above a certain level and in the vicinity of harbours. The occurrences of gastropod imposex and butyltin contamination in seawater, sediments, and green mussels from coastal waters of Thailand have been carried out since 1995 in order to assess the status of TBT contamination in the Thai coastal waters. Studies indicated that TBT contamination was a problem in areas with shipyards and repairing facilities or associated with elevated activities of ocean-going vessels or commercial shipping. The situation of TBT contamination in the Thai waters, however is improving; although several TBT hotspots in sediments still exist in the inner Gulf of Thailand and high levels of imposex are still being reported from the east coast of Phuket Island. Restrictions on the use of TBT antifouling paints are in the process of being implemented whereas Coastal Water Quality Standard for TBT has already been imposed. More data are needed on TBT levels in other marine organisms for further evaluation.

Key words: butyltins, tributyltin, anti-fouling, green mussels, dugongs, imposex, Gulf of Thailand

Introduction

Butyltin (BT) compounds have been used in industrial products such as polyvinyl chloride stabilizers, industrial catalysts, agricultural biocides, wood preservatives, and antifouling agents in paints since early 1970s. Tributyltin (TBT) has received much attention due to its wide application in antifouling paints and the resulting direct pollution of the estuarine and marine environment. TBT is an extremely effective antifoulant due to its broad spectrum toxicity and ability to persist on vessel hulls. The problem with TBT is that the threshold for harmful effects (hormone disrupter) is extremely low in certain mollusks. Chronic effects in oysters, mussels and crustaceans are observed at exposure levels of less than 1 μg TBT/L while the most sensitive species (dogwhelk snails, e.g. *Nucella lapillus* and *Buccinum undatum*) show sublethal effects at concentrations of only a few ng TBT/L or less (Svavarsson 2000, Jakobsen and Asmund 2000). The toxic effects are wide ranging and include imposex in shellfish, a reduction in shell growth in the mussel *Mytilus edulis*, and disruption of the ionic regulatory functions of fish. Effects have been observed in non-target organisms in non-target habitats worldwide (Alzieu et al. 1986, Bryan et al. 1986, Langston et al. 1990, Minchin and Minchin 1997, Swennen et al. 1997, Berge and Walday 1999, Yan and Yan 2003, Terlizzi et al. 2004, Roepke et al. 2005). Concerns over the serious ecotoxicological impacts of TBT to target and non-target marine organisms led to a ban on the application of TBT on boats shorter than 25 m in length in the USA, Australia, Japan and many EU countries (Evans et al. 1995, Gibson and Wilson 2003). However, TBT is still being used in antifouling paints applied to larger ships (>25 m in length) in most developed countries, and no restriction has been implemented on the usage in most Southeast Asian countries, including Thailand.

Thailand is one of the fastest growing developing countries in Southeast Asia. Thailand's population increased from 34 million in 1970 to 65 million by 2006. The rapid increasing population with associated industrialization and economic development in the coastal areas in the past two decades has resulted in construction and planning of many coastal development projects. Coastal waters have been subjected to pollution through the discharge of urban and industrial wastes. The increased numbers of shipbuilding yards, ship-repair services and marinas have imposed the potential risk for considerable antifouling biocides in the Thai estuarine and marine ecosystems. The prohibition of the use of TBT paints on vessels under 25 m in length, effective in France from 1982, in the United Kingdom from 1987 and more widely from the end of the 1980s and early 1990s, did much to improve the situation within marinas and sheltered harbours where use on leisure craft had predominated in many countries. Restrictions on the use of TBT antifouling paints have yet to be established in Thailand, but recently the subject has been receiving increasing attention. Here the existing data on the TBT concentrations in seawater, sediments and marine organisms in coastal areas of Thailand are reviewed in order to document the present status of TBT contamination in the Thai coastal waters.

TBT Concentrations in Seawater

TBT enters the water column through leaching and/or subsequent scraping of the hulls where it has caused numerous deleterious effects on non-target animals. TBT can be either dissolved or associated with particles smaller than 0.2 μm in diameter. The chemical half-life of TBT in water is approximately one week and is thought to be controlled by biological uptake and subsequent degradation. Under normal conditions, TBT may occur in the aquatic environment as various chemical species. Monobutyltin (MBT), dibutyltin (DBT), and inorganic tin are the main degradation products of TBT, resulting from biodegradation and photodegradation cleavage processes. Investigations concerning TBT in the coastal waters of Thailand started rather late due to lack of expertise. Advanced analytical methodologies or protocols, using GC/FPD or GC-MS, are required for the determination of ultratrace TBT and metabolites at the parts-per trillion levels in marine and estuarine waters. However, imposex incidences were reported to occur in *Murex* sp. and other gastropod genera from many coastal areas in the Gulf of Thailand (Swennen et al. 1996, 1997).

Lommetta (2001) conducted a baseline study of BT contamination in coastal water samples along the eastern part of the upper Gulf of Thailand in 2000, and reported TBT in the range from 43–223 ng/l. The highest value was found at Sattahip fishing boat pier and the lowest was at Pattaya recreational beach. At six fishing boat piers, concentrations were generally 69–223 ng/l; at three coastal mariculture areas, concentrations were 69–146 ng/l; at two commercial vessel traffic and berthing areas, concentrations were 113–144 ng/l.

However, the first extensive survey of TBT contamination in coastal waters was undertaken in 2003 as part of the Coastal Waters Monitoring Program by the Pollution Control Department (PCD) under the Ministry of Natural Resources and Environment. Seawater samples were collected from selected stations scattered through 23 coastal provinces of Thailand. The three year (2003–2005) results indicated that the concentrations of TBT in coastal water ranged from <2–7 ng/L in unexposed areas (such as recreational beach, fisheries with small boats and small scale aquaculture), and from 12–140 ng/l in areas with high boating activities especially in or near large harbours and major shipping traffic areas (PCD 2003, 2004, 2005).

Wattayakorn (2008) conducted BT analysis in coastal water samples from selected sites in the inner Gulf of Thailand (Figure 1). TBT concentrations were found to range from 2–39 ng/l and 3–30 ng/l for 2005 and 2006 sampling period, respectively. Although the majority of these measurements were generally below the proposed Thailand Coastal Water Quality Standard for TBT (10 ng/l), the levels still high enough to induce toxicological effects in sensitive species (Svavarsson 2000, Jakobsen and Asmund 2000). The highest TBT concentrations were found at Laem Chabang

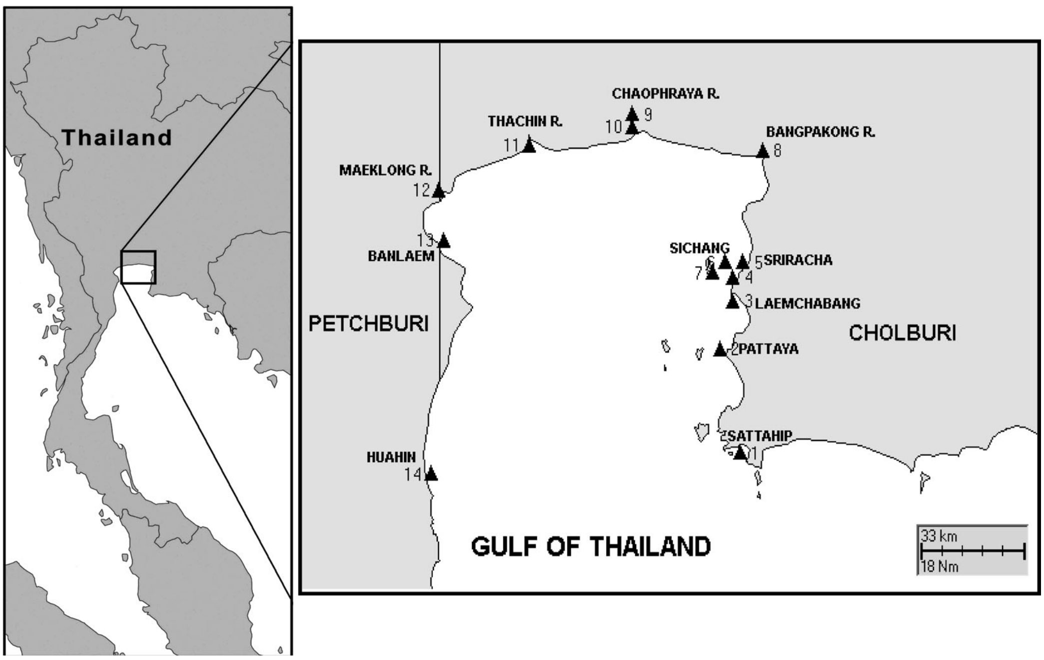

Fig. 1. Map of sampling stations along the inner Gulf of Thailand.

(average 27 ng/l), Ao Udom (average 34 ng/l) and Sriracha (average 25 ng/l) in Chonburi Province, where ocean-going vessels are usually anchored while waiting for their turn to berth. The high concentrations of TBT were likely due to leaching of TBT from antifouling paint used on the hulls of the moored vessels. The percentage proportion of TBT in ∑BTs for the water samples was in the range of 22 to 67%, whereas DBT and MBT ranged from 22 to 57% and 9 to 37%, with the average of 39%, 39% and 22%, respectively. The percentage proportion of [MBT]/[∑BTs] is used to determine whether the discharge is recent. Low percentages (3–10%) indicate recent discharge of TBT (Ko et al. 1995). The lowest percentages of [MBT]/[∑BTs] (9, 10 and 11%) were found at three shipping ports, Ao Udom, Sriracha and Laem Chabang, respectively, suggesting that the major source of TBT contamination in the inner Gulf is from antifouling paints used on ocean-going commercial vessels, rather than small boats.

TBT Concentrations in Sediments

The low solubility and high octanol/water partition coefficient (log K_{ow} is 3.54 for seawater) indicating that TBT partitions to the organic solvent and adsorbs strongly to particulate matter. Adsorption is considered to be an important mechanism for removal of organotins from the water column, however, desorption and sediment resuspension or uptake by benthic biota could also influence removal of organotin compounds from the aquatic environment. In sediments, TBT is generally degraded by bacteria into DBT and MBT which are less toxic forms of their parent compound. Considerable evidence suggests that TBT degradation kinetics in sediments is a much slower process than in the water phase, with half-lives on the order of years (Sarradin et al. 1991). Hence, sediments act as a relatively long-term repository for TBT and help to maintain its persistence in the aquatic environment. As a result, TBT in sediments has received increasing attention in recent years.

The first survey of BT contamination in sediments from the Thai coastal waters was conducted in 1995 (Kan-Atireklap et al. 1997a). Concentrations of BTs were reported in the range from 7 to 410 ng/g for MBT, 2 to 1900 ng/g for DBT and 4 to 4500 ng/g for TBT. The percentage proportion of TBT in ∑BTs was in the range of 21 to 91%, whereas DBT and MBT ranged from 3 to 36% and 5 to 59%, respectively. The presence of detectable TBT in sediments in all locations suggests its widespread contamination. Generally, TBT concentrations were in the range 4–81 ng/g dry weight in coastal mariculture sites, 7–93 ng/g at fishing boat piers, 9–880 ng/g at ports/fishing boat piers and 3,600–4,500 ng/g in offshore vessel harbours. The spatial distribution of BTs in sediments was apparently associated with boating activities.

Table 1. Concentration of butyltin compounds (ng/g dry weight) in sediments from the inner Gulf of Thailand (Harino et al. 2006)

Station	Location	MBT	DBT	TBT
1	Sattahip, Chonburi	25	38	72
2	Pattaya, Chonburi	10	8	5
3	Laem Chabang, Chonburi	59	106	284
4	Ao Udom, Chonburi	84	85	285
5	Sriracha, Chonburi	6	4	3
7	Sichang, Chonburi	37	20	34
8	Bangpakong River, mouth	293	77	28
9	Choa Phraya River, mouth	27	19	43
10	Chao Phraya River, mouth	3	6	10
11	Thachin River, mouth	158	368	1,246
12	Maeklong River, mouth	11	5	10
13	Ban Laem, Petchburi	13	5	12
14	Hua Hin, Prachuabkhirikhan	1	1	2

The high TBT concentrations were considered to be associated with elevated activities of far seas vessels or commercial shipping.

Lommetta (2001) analyzed sediment samples from 12 stations along the eastern part of the upper Gulf of Thailand in 2000, and reported TBT concentrations in surface sediments in the range 2–11 ng/g dry weight at fishing boat piers, 27–87 ng/g in coastal mariculture areas and 11–31 ng/g in commercial vessel berthing harbours.

In 2004, Harino et al. (2006) conducted a survey of BTs in surface sediments from selected stations around the inner Gulf of Thailand (Figure 1), and reported the concentrations of MBT, DBT and TBT in sediments in the range 1–292 ng/g, 1–368 ng/g and 2–1,246 ng/g dry weight, respectively. The higher concentrations of TBT in sediments were observed in stations at 3, 4, 8 and 11 (Table 1). Especially, the highest concentration of TBT (1,246 ng/g) was found at station 11 which is in the vicinity of shipyards with repair facilities, suggesting the deposit of the paint chips containing TBT to sediments. Contaminated sediments are therefore potential environmental reservoirs for TBT, and may continue to be a source long after the industrial use of TBT has been reduced. The percentage proportion of TBT in ∑BTs for the sediment samples was in the range of 7 to 70%, whereas DBT and MBT ranged from 17 to 33% and 9 to 74%, respectively. The percentage of TBT, DBT and MBT was on the average, 44%, 24% and 33% of the ∑BTs, respectively. The lowest percentage of [MBT]/[∑BTs] (9%) was found at the mouth of the Thachin River, suggesting a TBT "hot spot" for the inner Gulf. The ratio of TBT among BTs was over 60% in stations 3, 4 and 11 where TBT concentrations were high.

The concentrations of BTs in sediment from the inner Gulf of Thailand were shown to be reduced recently as compared to during 1994–1995. TBT concentrations in Thailand were higher than the TBT concentrations in north and central

Vietnam (Midorikawa et al. 2004) but similar to those in Malaysia (Sudaryanto et al. 2004).

TBT concentrations in Marine Organisms

The lipophilic nature of organometallic compounds such as TBT is often invoked as an explanation for the high partition coefficients and hence bioavailability of such compounds in marine organisms, from water and sediment. Butyltin compounds were detected in clams, mussels, oysters, fish and a wide range of marine wildlife.

Kan-Atireklap et al. (1997b) analyzed BT residues in green mussels (*Perna viridis*) collected in 1994 and 1995 from coastal waters of Thailand. BT compounds, such as TBT, DBT and MBT were detected in most mussel samples, ranging from 4 to 800 ng/g wet weight (as total). The composition of butyltin derivatives in green mussel was in the order of TBT>DBT>MBT. The concentration ranges were from <3–45 ng/g for MBT, 1–80 ng/g for DBT and 3–680 ng/g for TBT. The degree of TBT contamination in mussel tissues were rated as ‘low’ to ‘moderate’, except for a location in Kung Kra Baen, Chanthaburi province, with ‘heavy’ TBT contamination (680 ng/g). Relatively high concentrations of TBT were found in samples collected from areas with high boating activities and coastal aquaculture facilities.

Lommetta (2001) reported that TBT concentrations in the tissue of cockles (*Anadara granosa*), mussels (*Perna viridis*) and oysters (*Saccostrea* sp.) collected from mariculture sites in Chonburi Province were 4, 24 and 73 ng/g wet weight, respectively. The results were comparable to those reported in cockles and mussels collected from Peninsular Malaysia waters (Tong et al. 1996).

Harino et al. (2006) reported the concentrations of MBT, DBT, and TBT in green mussels (*Perna viridis*) from Thailand were in the range of 8–20 ng/g, 4–9 ng/g and 4–45 ng/g wet weight, respectively. The mean concentration of TBT in mussels was 16.2 ng/g, which was in the range of detected values from other Asian developing countries (Cambodia, China, India, Indonesia, Malaysia, Philippine and Vietnam) (Sudaryanto et al. 2002, Harino et al. 2008). The highest concentration was found at station 5 (Fig. 2) which is an aquaculture area, with heavy cargo shipping activities. Although the highest concentration of TBT in green mussels was found at station 5, the concentration of TBT in the sediments was lower than that at the other sites. In other words, TBT in the sediments was low, despite the higher concentration of TBT in water.

In addition to mussels, Harino et al. (2007) measured BT concentrations in organs and tissues of stranded dugongs (*Dugong dugon*) from the coastal waters of Thailand. Concentrations of BTs were in the range of 14–14,468 ng/g wet weight with the highest concentration in the livers, similar

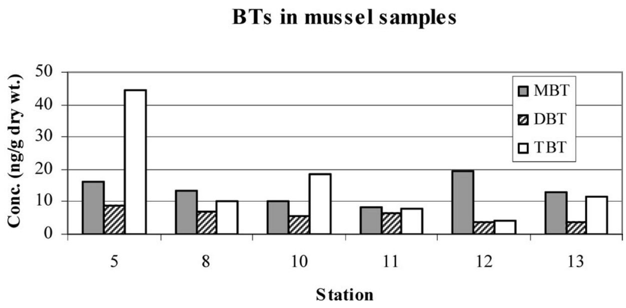

Fig. 2. Concentrations of butyltin compounds in mussel samples (Harino et al., 2006)

pattern to that found in other aquatic organisms. However, TBT was scarcely detected in the tissues and organs of dugongs, and MBT and DBT were the dominant compounds among the BTs. Comparison of BT concentrations in the tissues and organs of dugongs from the Gulf of Thailand with those from the Andaman Sea suggesting that BT contamination in the Gulf of Thailand was at similar levels to those in the Andaman Sea in 1998.

Occurrence of Gastropod Imposex

Imposex is the imposition of male sexual characteristics on female marine snails. Studies have shown that it is a very specific response to TBT exposure, although it is possible that imposex may also be triggered by other factors. Today imposex has been described in at least 118 species of neogastropods, belonging to 63 genera (Fioroni et al. 1991). Several studies have shown a significant correlation between the severity of imposex and the concentration of TBT in the sediment (Axiak et al. 1995), water (Stroben et al. 1992) or tissue of the gastropod (Gibbs et al. 1990). These findings have supported the assumption that TBT is undoubtedly a cause of imposex (Evans et al. 2000).

Surveys of gastropod imposex were undertaken in Thailand by Swennen et al. (1996, 1997) in the northern part of the Gulf of Thailand and in the southern part around Pattani Bay (related to distance from main shipping routes). The study was based on small sample sizes but it was the first report from the area. In the northernmost area gastropods sampled in the eastern part of the inner Gulf of Thailand, near the major shipping route to Bangkok and near oil terminals and a berthing place showed high imposex incidence (80–100%) in *Murex* sp. and other gastropod genera. The very low shipping density in the western part was reflected by no findings of imposex.

Bech (2002a) conducted more extensive imposex surveys of gastropods in Phangnga Bay on the west coast of Thailand from 1996 to 2000, where Kan-atireklap et al. (1997a) recorded the second highest concentration of TBT in the country from sediment samples collected at the deep sea port of Phuket Island (3900 ng/g dry weight). The incidence of imposex *Thais distinguenda* was found to increase significantly from 1996 to 2000, from an average of 22.5% in 1996 to 62.0% in year 2000, with some stations exhibited 100% imposex incidence since 1998. Increasing intensity of imposex was also observed for Thais bitubercularis, however the less sensitive species, i.e. *Morula musiva*, *Morula granulata*, *Morula margariticola* and *Thais rufotincta* only developed imposex at the three main areas of intense shipping activities. *T. bitubercularis* and *T. distinguenda* were recommended as indicators of TBT in Southeast Asia because of their sensitivity and wide distribution in the region. In some areas (Raja Island and Phi Phi Island), imposex incidence was found as a consequence of the establishment of a marina, and increasing yachting activities. The incidence of imposex was 100% at stations close to the yacht mooring sites and in the inner part of the bay, and declining towards the open sea (Bech 2002b). He suggested that TBT contamination was worsening due to lack of regulations prohibiting the use of TBT-based paints in Thailand. However, earlier studies have shown that imposex incidence must be used with caution. A major problem is that, while TBT is undoubtedly a cause of imposex, it is not the sole cause. Agents such as triphenyltin or nonylphenol can also cause it (Horiguchi et al. 1997, Evans et al. 2000).

Conclusions

Although information regarding the usage of BT compounds in Thailand is not available, the results presented in this paper indicate that TBT is a widespread problem and posing a threat to sensitive species in coastal areas of Thailand. Significant contamination of TBT has occurred in areas with dense shipping-related activities, such as commercial ports, dockyards and marinas. TBT "hot-spots" in seawater around the inner Gulf were found close to ocean-going commercial ports. Poor dockyard practices, allowing TBT-contaminated wastes, including paint flakes, to accumulate in sediments and left a legacy of hot-spots of contamination in some areas. In addition, areas close to marina developments and recreational boating, sensitive female gastropods were found to develop imposex which affecting the gastropod populations.

The planned phasing out of TBT, as recommended by the IMO, is likely to lead to a decline of TBT contamination in the Thai coastal waters and the improvement of gastropod imposex incidences. However, management effort would be best directed at addressing the continued input from ship maintenance facilities, which could include stricter controls on operations or a need for site remediation, and close monitoring of activities which could result in further inputs to the marine environment. The Pollution Control Department has recently imposed the Coastal Water Quality Standard for TBT in Thailand, while restrictions on the use of TBT antifouling paints are planning to be implemented soon. In addition, a greater monitoring and impact assessment of TBT from ports and marinas in Thailand should be continued in order to further detail the distribution and fate of TBT in the coastal waters. There is also a need to assess the potential impacts arising from TBT contamination. This should include both field studies of the ecotoxicological effects of TBT in coastal waters and laboratory toxicity testing on local marine organisms, to better understand and realize the potential threats from TBT contamination. However, because of the great stability of TBT in the sediments, it will probably stay in the coastal environment of Thailand for a while even after total banning.

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