Technical note

Butyltin in ballast water of merchant ships

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Abstract

The time series of analyzed values indicated that concentration of butyltin (BT) compounds in ballast water varied widely depending on the tanks, depths, and dates of sampling, ranging 4–93 ng l^{-1} during 7-day voyage. Variations in concentrations of BT in the two ballast tanks corresponded well with the level of BT contamination in different pumping sites of ballast waters. Concentrations of BT species detected in ballast water were rather consistent with those found in port waters and sediment of Taiwan and Japan. This study suggested that the problems concerning BT contamination were neither static nor localized.
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1. Introduction

A common and necessary practice for marine vessels is to pump substantial amount of water into the specifically designed ballast tanks to maintain structural integrity and adjust draft to enhance maneuverability and propulsion efficiency. A large containership, for example, may carry as much as 15,000 tons of ballast water (Talley, 2005).

It has been widely shown that transport of ballast water in ships is one of the most widespread mechanism by which nonindigenous aquatic species introduction occur (McDowell, 2002; Ruiz et al., 2001; Cohen and Carlton, 1998; US Coast Guard, 2001; US Coast Guard and US Department of Transportation, 2002; Matej and Marko, 2004). The same mechanism might also cause the introduction of chemical pollutants to other regions. These nonindigenous aquatic lives and pollutants might disrupt the local ecological system, when the ballast water transported from other regions was discharged.

According to the International Maritime Organization (IMO) (Oemcke and VanLeeuwen, 2003) an estimated 10 billion tons of ballast water are transferred around the globe each year. The transportation is not limited to international or cross-ocean shipping. The coastwise transport of ballast water can also spread chemical pollutants and aquatic nuisance species along the coastline (Ruiz et al., 2001). However, few have attempted to address the possible intake of chemical pollutants with ballast water and whether the discharge of ballast water is accompanied by the threat of chemical pollution to the reception regions.

Since early 1970s, tributyltin (TBT) has been used as antifouling agent in boat and ship paint applied to the hulls of vessels. The harmful effects of TBT released by antifouling paints were first documented in France Arcachon Bay at the end of 1970s (Alzieu, 2000). In addition to direct leaches from the ship hull, TBT can also be released into the environment in the form of paint wastes from sandblasting and hydroblast, which are the necessary maintenance procedures in dockyard. Due to the wide applications of antifouling paints, considerable amounts of butyltin (BT) compounds have entered estuarine and marine ecosystems (Hoch, 2001).

Even at very low concentrations (<10 ng l^{-1}), TBT can cause shell anomalies and failure of spat in oyster, impotence in neogastropods and gastropods (Horiguchi et al., 1998a, b; Hong et al., 2002), and immunological dysfunction in fish (Shimizu et al., 2003). At present regulations on TBT-based paints are not in conformity

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across the world. Some countries have no restrictions on the use and release of TBT antifouling paints, while others, including Japan, France, and several North Sea states, have more stringent restrictions.

The aim of the present study was to assess the current levels of BT compounds in the ballast water transported by merchant vessels and to provide baseline data for future efforts to decrease the threat of chemical pollutants through ship ballasting practice. The time series of concentration of BT compounds in ballast water was established to demonstrate variations in BTs in different ballast tanks and its correspondence with the level of BT contamination in international seaports in which ballast waters were pumped onboard.

2. Material and methods

2.1. Sample collection

2.1.1. Water samples

Water samples were collected through tank sounding pipe directly into 50-ml centrifuge tubes (polypropylene,

Table 1
Information of container ship M.V. KC on which ballast water was sampled

Delivery date	April 2000	Depth to main deck	13.5 m
Cargo capacity	1660 TEU[a]	Draught	9.94 m
Service speed	21 mile/h	Voyage course	SE Asia—Taiwan—Japan
Length overall	180 m	Gross tonnage	16,266 GT
Breadth	25 m	Net tonnage	8739 NT
Deadweight	22,494 MT	Total ballasting capacity	7663 m^3

[a]20 ft equivalent unit container.

graduations printed on outer surface), throughout the study. The centrifuge tube was loaded on a carrier fabricated from a 25.4 mm-diameter stainless-steel pipe. The depths of water sampling were measured at the same time.

Ballast water was sampled at intervals from the surface, half-depth, and bottom of ballast water column in the Wing Tank and the Heel Tank of M.V. KC (Table 1) during 7-day voyage (August 23–29, 2004) from Keelung Harbor, Taiwan to Tokyo Harbor, Japan (Fig. 1). The sites of ballast water intake in the Wing Tank were mainly near the exit of the Keelung Harbor, and the depth of ballast water was approximately 2.8 m at the time of sampling. The sites of ballast water intake in the Heel Tank were at the innermost of Tokyo Port. The depth of water was approximately 3.2 m at the time of sampling.

Water samples were also taken from Keelung Harbor and five other Japanese international seaports which include Tokyo, Yokohama, Kawasaki, Yakkaichi, and Nagoya. Three subsamples were taken from each water sample (about 100 ml/subsample). These water samples were acidified to pH 4.8 with acetic acid–sodium acetate buffer (1 M) immediately after sampling, and were frozen as quickly as possible and transferred to the laboratory where they were stored at −20 °C.

2.1.2. Sediment sample

Sediment samples (about 50 g) were collected from three locations around Keelung Harbor (Fig. 1). Sampling locations were chosen so as to provide sufficient representative coverage of the harbor. Samples were taken with an Ekman grab sampler (Wildco Wildlife, Saginaw, MI, USA) installed on a winch set of a research boat. Sediment samples were immediately frozen and transferred to the laboratory for storage and later extraction.
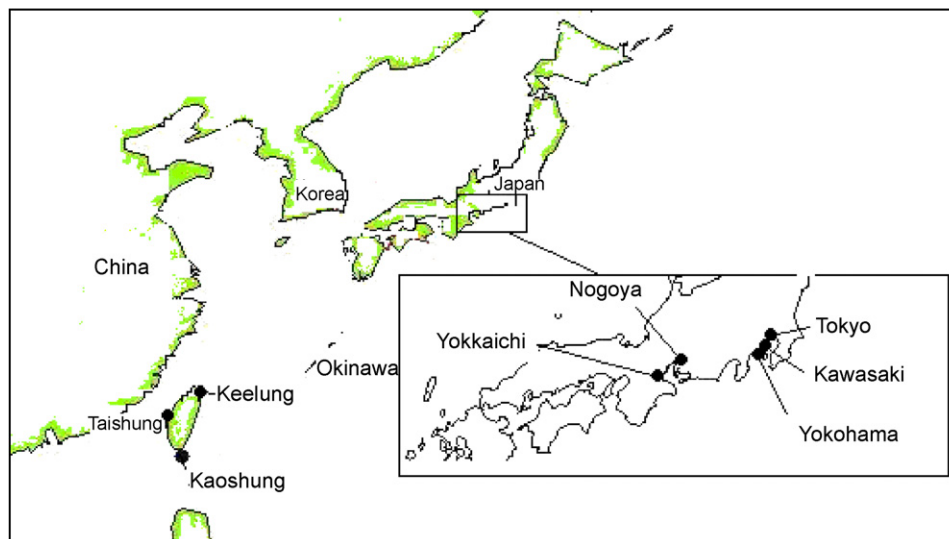

Fig. 1. Locations of international seaports M.V. KC have visited and ballast waters have been taken during its 7-day voyage, ● marks the sampling points of the port waters. Separated file: Fig. 1 mapTaiwanJapan.

2.2. Analytical procedures for BT compounds

BT compounds in the ballast water and harbor water samples were analyzed as described in a previous report (Hsia and Liu, 2004). Briefly, the analytical procedure consisted of two steps: (1) solid phase microextraction (SPME) procedure (Hsia and Liu, 2003) is used for sodium tetraethylborate derivatization and extraction to the fiber; (2) quantitative determination of BT compounds by a Dani GC 1000 gas chromatograph equipped with a column (HP−5, $30\,m \times 0.25\,mm$ i.d. $\times 0.25\,\mu m$ film thickness, Hewlett Packard, USA) and a flame photometric detector fitted with a 610 nm optical fiber. For quantification, internal standard tripropyltin chloride was employed and added to the sample prior to extraction. All experiments were conducted in triplicate.

Concentration of BT compounds in water samples and in sediment samples were expressed as $ng\,l^{-1}$ and $\mu g\,g^{-1}$ on a wet weight basis, respectively. Recoveries of the BT compounds from spiked-in-sea water ($50\,g\,l^{-1}$, $N = 6$) and in sediment samples ($50\,\mu g\,g^{-1}$, $N = 6$) ranged 95–111% and 92–102%, respectively. The method detection limit of the BT compounds in seawater and sediment ranged 3.5–$3.8\,ng\,l^{-1}$ and 2.2–$4.2\,\mu g\,g^{-1}$, respectively.

3. Results and discussion

3.1. Concentration of BTs in the wing tank of M. V. KC

The concentrations of TBT, dibutyltin (DBT), monobutyltin (MBT), and sum of TBT, DBT, and MBT (ΣBT) in ballast water sampled from the top-, middle-, and bottom-layer of the Wing Tank of M.V. KC during 7-day voyage are presented in Fig. 2.

BTs (54–$94\,ng\,l^{-1}$) were detected in all ballast water samples from the Wing Tank of M.V. KC. Their concentrations varied widely depending on the depths and dates of sampling, ranging 17–$53\,ng\,l^{-1}$ for TBT, 14 to $48\,ng\,l^{-1}$ for DBT, and 8 to $17\,ng\,l^{-1}$ for MBT (Fig. 2). The mean TBT concentration ($29\,ng\,l^{-1}$) was higher than those of DBT ($25\,ng\,l^{-1}$) and MBT ($12\,ng\,l^{-1}$). The high concentration of BTs in the water samples lead to the suspicion that some of the intake sites of the ballast water in Wing Tank might be contaminated with TBT. As most of the ballast water in Wing Tank was from Keelung Harbor, it is reasonable to infer water in that harbor was the source of problem, but it might also come from any other harbors M.V. KC had visited prior to this voyage.
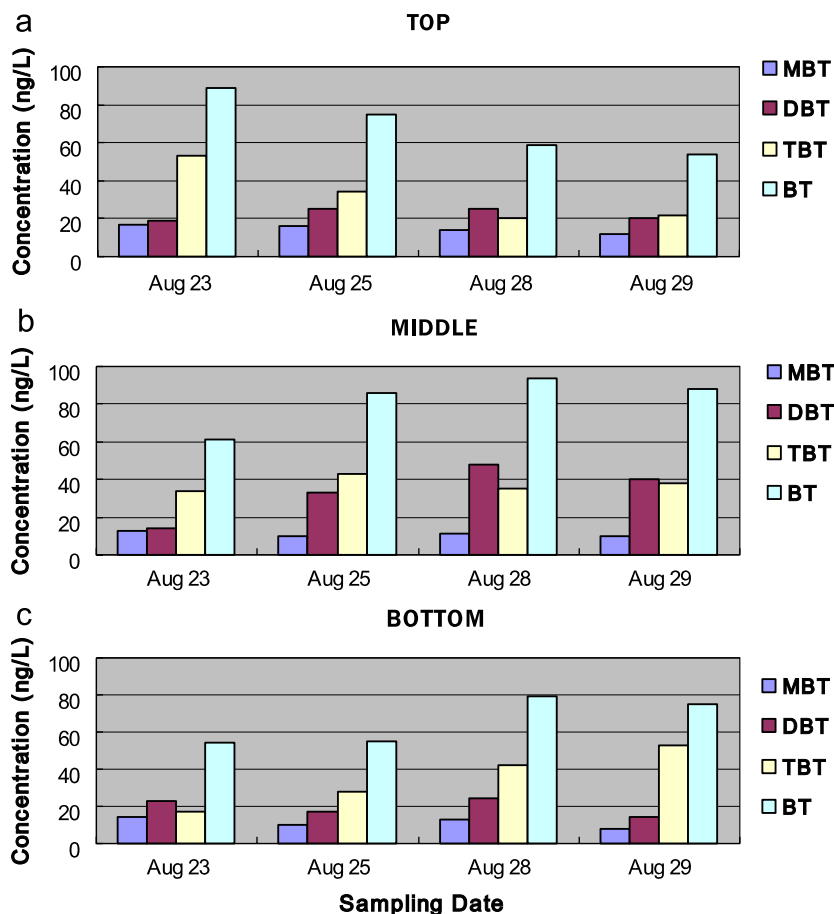

Fig. 2. Concentrations of TBT, DBT, MBT, and ΣBT (sum of TBT, DBT, and MBT) in ballast water sampled from the top, middle, and bottom layer of the Wing Tank of M.V. KC during the 7-day voyage.

In Fig. 2, it is interesting to note that TBT concentrations in the ballast water from the top layer of Wing Tank decreased (53–22 $ng\,l^{-1}$) following the sampling dates, but the concentrations of TBT in the ballast water from the bottom layer increased (16–53 $ng\,l^{-1}$). TBT concentrations in the ballast water from the middle layer were increased (within 34 and 41 $ng\,l^{-1}$) in the first 2 days then decreased slightly in the coming 4 days. Similar phenomena were seen in the concentrations of ΣBT. ΣBT concentration in the ballast water from the top layer of Wing Tank decreased (89–54 $ng\,l^{-1}$) throughout the sampling dates, but ΣBT concentration in the ballast water from the bottom layer increased (54–79 $ng\,l^{-1}$) in the first 5 days and then decreased next day. ΣBT concentrations in the middle layer of water column were also increased (within 61 and 94 $ng\,l^{-1}$) in the first 4 days then decreased slightly next day. In marine environment, BT compounds released from the antifouling paint on ship hull into water could be rapidly adsorbed to suspended organic matter and deposit (Michelsen et al., 1996; Barakat et al., 2001; Tolosa and Readman, 1996). Since the intake site of ballast water in the Wing Tank was mainly near the exit of the Keelung Harbor in the beginning of the voyage, the significant variations in concentrations of TBT and ΣBT in water columns of Wing Tank in response to the sequence of depths and sampling dates (Fig. 2) during the 7-day voyage could be attributed to settlement after BTs adsorption to particulates in the water column.

The slight decrease of ΣBT in the bottom layer of the ballast water column could also be the results of biodegradation since degradation of BT compounds is known to occur in water column (Kannan and Falndysz, 1997; Kröning and Rosenstock, 1989; Tolosa et al., 1996; Evans, 1999). Other factors, such as resuspension and desorption might also influence the concentration of BT compounds in the water column (Evans, 1997; IMO, 2005; Guruge and Tanabe, 2001; Kan-atireklap et al., 1997; Kuo and Shao, 1999). These latter explanations, however, require further investigation and are beyond the scope of this study.

3.2. Concentration of BT in the heel tank of M.V. KC

The concentrations of TBT, DBT, MBT, and ΣBT in ballast water sampled from various depths of Heel Tank of M.V. KC during the voyage are presented in Fig. 3. BT compounds (43–172 $ng\,l^{-1}$) were detected in all ballast water sampled from the Heel Tank. Their concentrations

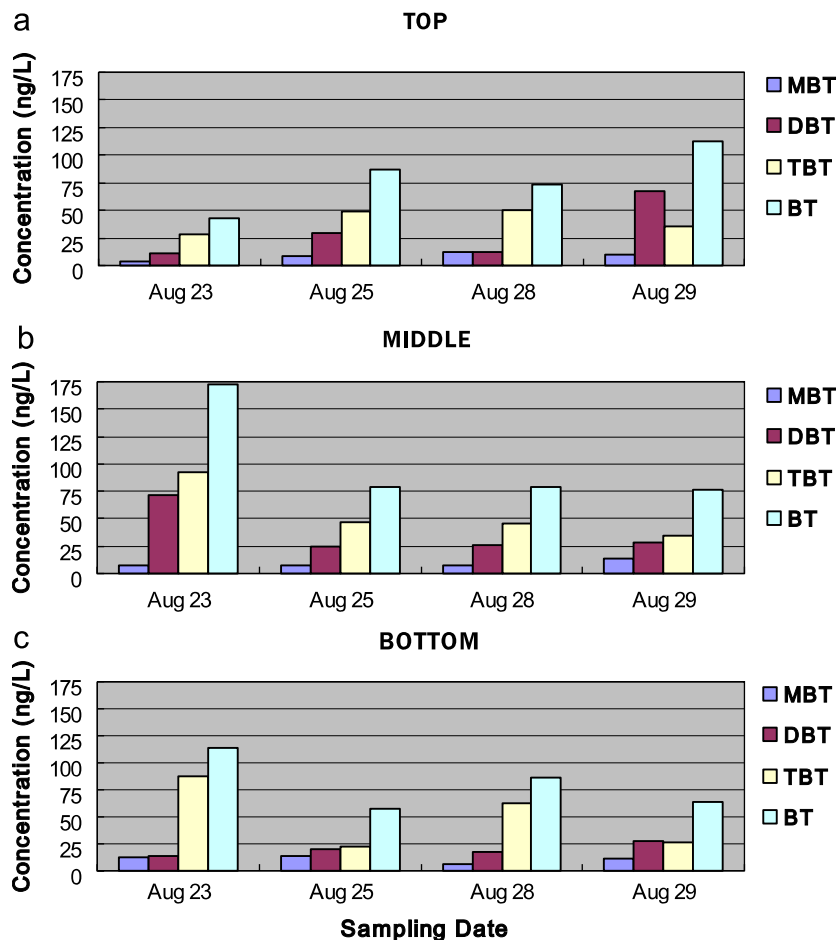Fig. 3. Concentrations of TBT, DBT, MBT, and ΣBT (sum of TBT, DBT, and MBT) in ballast water sampled from the top, middle, and bottom-layer of the Heel Tank of M.V. KC during the 7-day voyage.

also varied widely depending on the depths and dates of sampling, ranging 11–71 ng l^{-1} for MBT, 4–14 ng l^{-1} for DBT, and 23–93 ng l^{-1} for TBT (Fig. 3). In most cases, similar to that in the Wing Tank (Fig. 2), the mean TBT concentration (49 ng l^{-1}) was markedly higher than those of DBT (10 ng l^{-1}) and MBT (9 ng l^{-1}). The high concentrations of BTs in Heel Tank lead to the suspicion that some of the intake sites of the ballast water might be contaminated with TBT. As almost all of the ballast water in Heel Tank was either from Keelung Harbor or Japanese harbors, it is reasonable to infer water in those harbor were contaminated with TBT.

In Fig. 3, TBT concentrations in the top layer of ballast water column in Heel Tank increased (within 27 and 49 ng l^{-1}) in the first 2 days and remained at the same concentration for 2 days, and then decreased to 34 ng l^{-1} in the following 2 days. DBT concentrations fluctuated between 10 and 30 ng l^{-1} in the first 6 days, however, the concentration jumped to 63 ng l^{-1} on the 7th day. In most cases MBT concentrations were at low level (<10 ng l^{-1}). TBT concentrations in the middle layer of ballast water column in Heel Tank was as high as 93 ng l^{-1} in the first day, however, then decreased to 32 ng l^{-1} in the following sampling dates. DBT concentration decreased from 64 to 22 ng l^{-1} in the earlier 5 days and then increased to 23 ng l^{-1}. TBT concentrations in the bottom of ballast water in Heel Tank decreased from 80 to 24 ng l^{-1} in 2 days and then increased to 63 ng l^{-1}, and decreased to 25 ng l^{-1} in following day. Similar trend can be seen in the concentrations of ΣBT in the ballast water in the Heel Tank.

The trend of variations in the concentrations of TBT and ΣBT found in the Heel Tank were quite different from those found in the Wing Tank. Both TBT concentrations and the average TBT/DBT ratios in the analyzed ballast water in Heel Tank (Fig. 3) were markedly higher than those in the Wing Tank (Fig. 2). The intake sites of ballast waters in the Heel Tank were mainly at the innermost of Tokyo Harbor, while the intake sites for the Wing Tank

were near the exit of the Keelung Harbor. Thus the difference found in two different ballast tanks may be attributed to the higher level of TBT contamination in the innermost ballasting sites of M.V. KC in Tokyo Harbor.

3.3. BT concentration in water samples collected from Taiwan and Japanese harbors

Ballast water was extracted from both Wing Tank and Heel Tank in this study. Among various BT species, TBT was the most predominant at all depths and dates of sampling, with overall ranges 33–77% and an average of 53%. The high concentrations of TBT and relatively low concentrations of DBT and MBT in the ballast waters suggest that degradation of TBT was minor in ballast waters during the 7-day voyage. The ballast waters in Wing Tank (2.8 m) and Heel Tank (3.2 m) were virtually confined under dark and anaerobic conditions, little photodegradation and biodegradation of TBT can be assumed. Thus TBT concentrations in the ballast water reflected TBT contaminations in the areas where ballast water were taken.

Since it is common practice for a container ship to adjust its draft and trim in each ballasting process, by pumping unequal amount of water to partially fill ballast tanks from separate sources, it is difficult to pinpoint the sources of chemical contents in ballast water. However, this study demonstrates the potential of TBT transportation through ballast water carried by merchant cargo ships as Table 2 summarizes the results from M.V. KC (including five Japanese ports and one Taiwanese port) and other previous reports about BT contamination in marine environment of Asian coasts (Guruge and Tanabe, 2001; Kan-atireklap et al., 1997; Kuo and Shao, 1999; Shim et al., 2005; Murai et al., 2005, Lee, 2003; Harino et al., 1997).

BT concentrations in the water samples of five international seaports where ballast water was being pumped, ranged 15–56 ng l^{-1}. Variations in concentrations of BTs in the two ballast tanks (32–71 ng l^{-1} in Wing Tank;

Table 2
Comparison of organic-tin concentration around Asian international ports

Location	Sample type	TBT	DBT	MBT	Reference
Coast, Korea	Bivalves	48–2800 ng Sn/g	170–290 ng Sn/g	122–197 ng Sn/g	(Shim et al., 2005)
West coast, Japan	Water	9.0±7.0 ng l^{-1}	3.3±3.0 ng l^{-1}	8.2±9.2 ng l^{-1}	(Murai et al., 2005)
Keelung, Taiwan	Water	58–260 ng l^{-1}	ND	ND	(Lee, 2003)
Tokyo, Japan	Water	36±12 ng l^{-1}	14±7 ng l^{-1}	18±9 ng l^{-1}	This study
Yokohama, Japan	Water	35±15 ng l^{-1}	13±16 ng l^{-1}	46±11 ng l^{-1}	This study
Kawasaki, Japan	Water	12±7 ng l^{-1}	14±7 ng l^{-1}	11±4 ng l^{-1}	This study
Yakachi, Japan	Water	22±16 ng l^{-1}	14±11 ng l^{-1}	11±10 ng l^{-1}	This study
Nagoya, Japan	Water	26±27 ng l^{-1}	15±7 ng l^{-1}	22±4 ng l^{-1}	This study
[a]Hopindao, Taiwan	Sediment	126–254 μg g^{-1}	19–290 μg g^{-1}	81–183 μg g^{-1}	This study
[a]Bachiman, Taiwan	Sediment	289–363 μg g^{-1}	98–456 μg g^{-1}	120–226 μg g^{-1}	This study
[a]Hseiho, Taiwan	Sediment	180–282 μg g^{-1}	118–288 μg g^{-1}	36–207 μg g^{-1}	This study
Wing ballast tank, M.V. KC	Water	17–53 ng l^{-1}	14–48 ng l^{-1}	8–17 ng l^{-1}	This study
Heel ballast tank, M.V. KC	Water	23–93 ng l^{-1}	11–71 ng l^{-1}	4–14 ng l^{-1}	This study

[a]Within 15 km from Keelung Port.

$31–73\,\mathrm{ng}\,\mathrm{l}^{-1}$ in Heel Tank) corresponded well with the level of BT contamination in different pumping sites of ballast waters.

Significant concentrations of BT compounds were also found in sediment and water samples from three Taiwanese international ports, Keelung, Taichung, and Kaohsiong (Murai et al., 2005, Lee, 2003; Harino et al., 1997) where M.V. KC has been moored most. All the Taiwanese and Japanese harbors M.V. KC had visited are busy international ports. Frequent release of ballast water could lead to the release of BT compounds into the water column.

To alleviate the threat of TBT coating to their aquatic environments, most developed countries have their own regulations in restricting the usage of TBT on the hull of certain types of vessel since early 1980s (Evans, 1997, 1999). In October 2001 the IMO Diplomatic Conference passed a global treaty on the 'Control of Harmful Antifouling Systems on Ships' to ban the application of organotin antifoulants by 2003 (IMO, 2005). Nevertheless, virtually all the regulations on the use of TBT paints have not been applied to the ballast tanks (IMO, 2005). Since the average $\mathrm{TBT}/\Sigma\mathrm{BTs}$ ratio of the analyzed ballast water (0.53) in this study was higher than those of the analyzed values in port water (0.44), it is rational to suspect that TBT in the antifouling paint coated in the ballast tank might contribute to the increase of TBT concentration in the ballast waters.

Among water sampled from five Japanese ports in this study (Table 2), high total concentrations of BT ($94\,\mathrm{ng}\,\mathrm{l}^{-1}$ in average, with highest $141\,\mathrm{ng}\,\mathrm{l}^{-1}$ in Yokohama) were found in enclosed international ports with poor water turnover. TBT ($22–56\,\mathrm{ng}\,\mathrm{l}^{-1}$, $41\,\mathrm{ng}\,\mathrm{l}^{-1}$ in average) was detected in port waters and these results indicated that BT compounds were widespread contaminants in this region.

Despite a declining trend of TBT contamination observed extensively in the early 1980s after the restrictions on the use of TBT (Kan-atireklap et al., 1997; Horiguchi et al., 1998a, b), there were still high levels of BTs in marine organisms and/or waters of countries including Japan (Takeuchi et al., 2004a, b; Higashiyama et al., 1991). Higashiyama et al. (1991) found relatively high concentrations of organotin compounds persisting in blue mussels of Tokyo Bay. Murai et al. (2005) updated the status of BT pollution in Japan. Since Japan had banned the usage of TBT on all coastwise vessels and aquaculture facilities more than a decade ago, the spatial distribution of BT compounds in these port waters probably were released from the illegal usage of TBT (Takeuchi et al., 2004a, b), the local deposited contaminant in sediment (Michelsen et al, 1996), and/or contaminants derived from intensive commercial shipping activities (Sakai et al., 2003). The mean TBT concentration among the seawater sampled in this study (Table 2) may indicate that TBT was still a potential hazard to international port waters of Japan, and further other countries, through ballast water.

In this study, the container ship M.V. KC in this study has a gross tonnage, net tonnage, and total ballasting capacity of 16,266, 8739, and $7663\,\mathrm{m}^3$ (Table 1), respectively. Based upon the results in this study, on assuming an average TBT concentration of $50\,\mathrm{ng}\,\mathrm{l}^{-1}$ in ballast water, it yields $190\,\mathrm{g}$ of TBT input to port water in every visit by a container ship of similar type to M.V. KC, assuming 50% of the capacity of ballast water has to be pumped overboard for loading. In other words, a typical international port such as Keelung with 15,867 container ships and 24,198 ships of other types visiting in 2004, it can yield a rough estimate of $176 \times 10^6\,\mathrm{m}^3$ of ballast water or $4,400\,\mathrm{g}$ of TBT input to neighboring coast water annually.

This study demonstrates that the media of worldwide transportation of TBT is not limited to hull and other exterior portion of ships. Originating from inside of ships, ballast water may act as a vector of much greater capacity and contribute to the re-buildup of TBT contamination in estuaries, international seaports, and coastal regions. The alleviation of the problems rely on guidance in ballasting practice which can be achieved through in conjunction with those well-developed and documented guidelines for ballast water management (IMO, 1997, 2005). A comprehensive monitoring of BT compounds in ballast water on board of various types of ships and the assessment of the potential for its impact on aquatic environment can be the next step to take.

Acknowledgments

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