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## High Tributyltin Contamination in Sediments from Ports in Indonesia and Northern Kyushu, Japan

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Sediment samples were collected from 20 stations in the port areas around three cities (Jakarta, Bali and Manado) in Indonesia in 2011 and from 9 stations in the port areas in northern Kyushu, Japan in 2010. High concentrations of tributyltin (TBT) were found in sediments collected in front of the shipyard in Bitung port, Manado (4.25  $\mu\text{g/g}$  wet weight) and in Benoa port, Bali (0.21  $\mu\text{g/g}$  wet weight). High TBT concentrations were also detected in sediments from next to the shipyard in Hakata port (0.63  $\mu\text{g/g}$  wet weight), Fukuoka, Japan. These results suggest that ports might be major sources of TBT pollution in Indonesia and Japan, even though TBT usage has been regulated in Japan since 1990.

**Key words:** hot spot, port, pollution, sediment, tributyltin

### INTRODUCTION

Tributyltin (TBT) has been widely used as an anti-fouling agent in marine environments since the early 1960s (Hugget *et al.*, 1992), having been applied to boat hulls and to marine aquaculture equipment. Because of its high toxicity of TBT to aquatic organisms (Clark *et al.*, 1988), the use of TBT has been regulated worldwide since the 1980s. The International Maritime Organization (IMO) imposed a worldwide ban on TBT in 2003 (Champ, 2000). However, TBT is still detected in coastal sediments in the world (Antizar-Ladislao, 2008). Around northern Kyushu, Japan, the concentrations of TBT have been found to range from 0.005 to 0.094  $\mu\text{g/L}$  and 0.007 to 1.1  $\mu\text{g/g}$  dry weight (dw) in seawater and sediments, respectively (Inoue *et al.*, 2002), from 0.008 to 0.135  $\mu\text{g/g}$  in bivalves (Inoue *et al.*, 2006), and from 0.0014 to 0.19  $\mu\text{g/mL}$  in blood from marine fish (Miki *et al.*, 2011). Contamination of sediment and aquatic organism by TBT still occurred in coastal areas around northern Kyushu. These results suggest the existence of TBT hotspots that continue to release TBT into the wider environment.

The Indonesian archipelago consists of many islands, and, by area, is 75% water. Many people in Indonesia use boats, and boats are suspected of being a major source of TBT contamination. The use of TBT is not yet regulated in Indonesia, even though contamination of the Indonesian environment by butyltin compounds has been reported (Sudaryanto *et al.*, 2005; Midorikawa *et al.*, 2004). Total butyltin concentrations in sediments (dw), bivalves wet weight (ww), and fish (ww) of 0.023–

0.32  $\mu\text{g/g}$ , 0.005–0.064  $\mu\text{g/g}$ , 0.00041–0.019  $\mu\text{g/g}$  and 0.209–0.425  $\mu\text{g/g}$  have been found in the major harbors of Jakarta, Surabaya, Medan and Bitung Manado respectively (Rumengan *et al.*, 2008; Harino *et al.*, 2012). Shipyards are suspected of being hot spots in ports and harbors. Extremely high TBT concentrations (2837  $\mu\text{g/kg dw}$ ) were detected in surface sediment collected from Hong Kong port (Cheung *et al.*, 2003) and a Sn concentration of 36.3  $\mu\text{g/g}$  was found at a site in front of a dry dock and near surface runoff drainage point in a Korean shipyard (Kim *et al.*, 2011). There is currently little information on TBT concentrations in sediments in and near Indonesian shipyards.

In this study, we measured TBT concentrations in sediments collected from ports in Indonesia, where no regulations for the use of TBT and dibutyltin (DBT) have been implemented, and in sediments from ports in Japan, where TBT usage has been regulated since 1990.

### MATERIALS AND METHODS

#### Sediment sampling

Three areas of Indonesia, Manado, Bali and Jakarta were selected as sampling areas, and samples were collected in January 2011. The sampling sites in the north of Sulawesi Island were Manado port (Fig. 1A, Stations 1–3), marina in Manado port (Fig. 1A, Stations 4) and Bitung port (Fig. 1B, Stations 5–8). The sampling sites on Bali Island were in Benoa port (near Denpasar) (Fig. 1C, Stations 9–11). The sampling sites on Java Island were in Jakarta Bay, and Tanjung Priok port (Fig. 1D, Stations 12–14), marina in Ancol (Fig. 1D, Stations 15–17) and Sunda Kelapa port (Fig. 1D, Stations 18–20). Sediment samples (5 cm deep) were collected using an Eckman grab sampler (Rickly hydrological company, Mississippi, US). Samples from each site were pooled and stones were removed. The samples were subsequently transferred to 50 mL polyethylene tubes and kept at  $-30^{\circ}\text{C}$  during transportation. Japanese sediment sam-

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ples were collected from Hakata port in Hakata Bay (Fig. 1E, Stations 21–23) and Imari port, Imari bay (Fig. 1F, Stations 24–29) in September 2010. These sediment samples (5 cm deep) were collected using a core sampler (Gravity Core Sampler, Rigo-sha, Tokyo, Japan) and stored at  $-30^{\circ}\text{C}$ .

### Reagents

Tributyltin chloride- $\text{d}_{27}$  (TBTCl- $\text{d}_{27}$ ) and dibutyltin chloride- $\text{d}_{18}$  (DBTCl- $\text{d}_{18}$ ), used as internal standards, were purchased from Hayashi Pure Chemical Industries, Ltd. (Osaka, Japan). Sodium tetraethyl borate  $[(\text{NaB}(\text{C}_2\text{H}_5)_4)]$  and Florisil were obtained from Wako Pure Chemical Industries, Ltd. (Osaka, Japan). All organic solvents were of pesticide residue analytical grade. Other reagents used were of analytical grade.

### Analytical procedure for butyltin

The concentrations of butyltin compounds in the samples were analyzed following the method published by Iwamura *et al.*, (2000) with slight modifications. After washing all glassware with hexane and acetone, 1 g of each sample was spiked with 1  $\mu\text{g}$  each of the TBTCl- $\text{d}_{27}$  and DBTCl- $\text{d}_{18}$  internal standards, in a centrifuge tube. Then 10 mL of a 1:1:1 mixture of 1 M hydrobromic acid in methanol, ethyl acetate and deionized water was added. The tube was shaken for 30 min, and then centrifuged for 5 min at 3000 rpm about  $1500\times g$  in our centrifugation. The supernatant solution was separated and the sample was extracted twice with 10 mL of 3:2 ethyl acetate:hexane solution, with 10 min shaking followed by centrifuging for each extraction. The organic layers obtained were concentrated using a Turbo Vap II (Zymark, Hopkinton, MA, USA) with  $\text{N}_2$  gas flushing at  $35^{\circ}\text{C}$ , to 1 mL and made up to 10 mL with hexane. The sample was ethylated by adding 0.5 mL of a 5% sodium tetraethyl borate solution, to derivatize the target organotin compounds, and 10 mL of deionized water was added. The mixture was then shaken for 30 min. 10 mL of 1 M potassium hydroxide solution was added, to saponify any fat, and the sample was shaken for 60 min, then centrifuged at 3000 rpm for 5 min. The hexane layer was collected by pipette and concentrated before being cleaned up using a Florisil-packed chromatography column, with the organotin compounds being eluted with hexane. The resulting solution was concentrated again using the Turbo Vap II.

Butyltin concentrations were measured using an Agilent 6890 gas chromatograph (GC) (Avondale, PA, USA) equipped with an Agilent 5973 mass spectrometer (MS) operated in EI+ mode. The GC column was a 30 m  $0.21\text{ mm}$  id fused silica capillary column (HP-5MS, Agilent J & W, CA, US). The carrier gas was helium (Zero-U grade; flow rate:  $1.2\text{ mL/min}$ ). The GC temperature program was:  $60^{\circ}\text{C}$  for 2 min,  $20^{\circ}\text{C/min}$  to  $130^{\circ}\text{C}$ ,  $10^{\circ}\text{C/min}$  to  $210^{\circ}\text{C}$ ,  $5^{\circ}\text{C/min}$  to  $260^{\circ}\text{C}$ , then  $10^{\circ}\text{C/min}$  to  $300^{\circ}\text{C}$  (held for 2 min). The injector, interface, and ion source temperature were  $270^{\circ}\text{C}$ ,  $280^{\circ}\text{C}$ , and  $230^{\circ}\text{C}$ , respectively. The ionization voltage was 70 eV. Ions of DBT, DBTCl- $\text{d}_{18}$ , TBT and TBTCl- $\text{d}_{27}$  ions were moni-

tored at  $m/z$  of 261, 279, 263 and 318, respectively.

The accuracy of the TBT and DBT analysis technique was checked using the certified reference materials (CRMs) NIES-CRM No.11 (fish tissue) and NIES-CRM No.12 (marine sediment) (National Institute for Environmental Studies, Tsukuba, Japan). The butyltin concentrations were calculated from the ratio between the butyltin peak and the appropriate butyltin Cl internal standard peak areas, and are expressed as  $\mu\text{g/g}$  wet weight (ww). All samples were analyzed in triplicate. A TBT recovery test was performed ( $n=3$ ) with sediment samples containing an undetectable level of TBT spiked with 1  $\mu\text{g}$  of TBTCl.

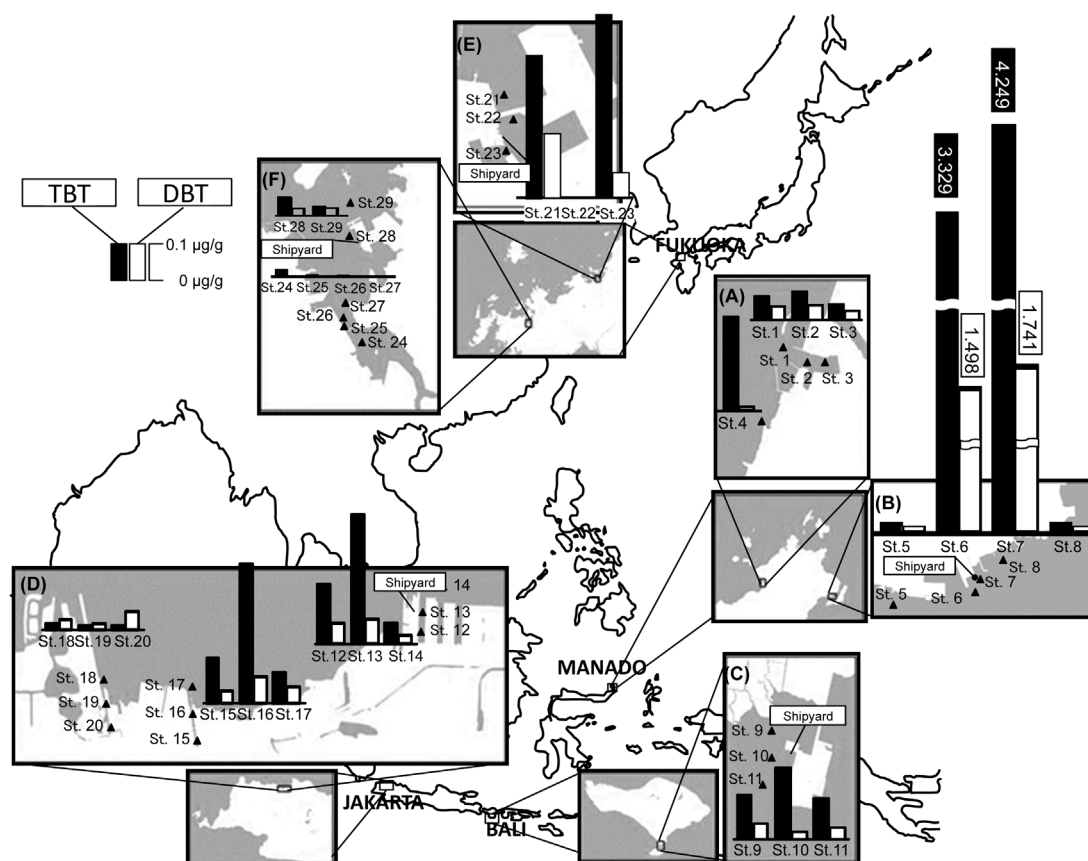
The TBT concentrations detected in the CRMs were  $1.17\mu\text{g/g}$  in NIES-CRM No. 11 (certified concentration  $1.3\mu\text{g/g}$ ) and  $0.16\mu\text{g/g}$  in NIES-CRM No.12 (certified concentration  $0.19\mu\text{g/g}$ ). Therefore, we concluded that our detected TBT concentrations agreed with the NIES-CRM certified concentrations.

## RESULTS

TBT and DBT were detected in the sediment samples from all 20 stations in Indonesia (Manado, Bali, Jakarta, Fig. 1A–D) with TBT concentrations of  $0.007\text{--}4.25\mu\text{g/g}$  ww and DBT concentrations between  $0.009$  to  $1.74\mu\text{g/g}$  ww. In the Jakarta Bay samples (Fig. 1D), the highest TBT concentration was found in a sample taken from marina in Ancol (Station 16,  $0.33\mu\text{g/g}$  ww), and the next highest concentration was found in a sample from close to the shipyard in Tanjung Priok port (Station 13,  $0.32\mu\text{g/g}$  ww). Low TBT concentrations were detected in samples from the other sampling sites. TBT was found at concentrations between  $0.007\text{--}0.015\mu\text{g/g}$  ww in samples from Sunda Kelapa port. (Fig. 1D, Stations 18–20), where DBT concentrations were higher than TBT concentrations. The highest TBT concentration ( $0.21\mu\text{g/g}$  ww) in the Benoa port, Bali (Fig. 1D) area was found at Station 10, which is very closed to the shipyard.

The highest TBT concentrations in the Manado area samples were found in samples from Bitung port ( $4.25\mu\text{g/g}$  ww, Fig. 1 B, Station 7, followed by Station 6,  $3.44\mu\text{g/g}$  ww), these were close to the shipyard. Low TBT concentrations were found at the other Manado sampling sites. TBT was found at just  $0.04\mu\text{g/g}$  ww in a sample from Station 3 in Manado port (Fig. 1 A).

The highest TBT concentration found in the Japanese samples was from sediment collected from Station 23 ( $0.63\mu\text{g/g}$  ww), which is located closed to the shipyard in Hakata port, and the next highest concentration was in sediment from Station 21 ( $0.49\mu\text{g/g}$  ww). TBT and DBT were not detected ( $<0.005\mu\text{g/g}$ ) at Station 22. The highest DBT concentration in the Japanese samples ( $0.21\mu\text{g/g}$  ww) was also found at station 21, and next highest was found at Station 23 ( $0.08\mu\text{g/g}$  ww). TBT and DBT were only detected in Imari port at Station 28, which is located in front of the shipyard, with a TBT concentration of  $0.06\mu\text{g/g}$  ww and a DBT concentration of  $0.02\mu\text{g/g}$  ww, TBT and DBT were not detected ( $<0.005\mu\text{g/g}$ ) at Station 27.



**Figure 1.** Map showing sample site locations and the concentrations of butyltin (TBT and DBT) in sediment samples from Indonesia [(A) Manado port, (B) Bitung port, (C) Benoa port, Bali and (D) Tanjung Priok port, Marina in Ancol, Sunda Kelapa port] and Fukuoka, Japan [(E) Hakata port, (F) Imari port].

## DISCUSSION

High TBT concentrations were detected in sediment samples collected from ports in Indonesia and Japan. These results suggest that ports might be major sources of TBT pollution in both Indonesia and Japan, even though TBT usage has been regulated in Japan since 1990.

The highest TBT and DBT concentrations in the Indonesia samples were found in Bitung port, Manado, in north Sulawesi, which is the fourth biggest port in Indonesia.

Imposex symptoms have been found in biota from coastal areas of the island of Ambon (eastern Indonesia), and were presumed to have been caused by TBT compounds leaching from the anti-fouling paints used on boat hulls (Ellis *et al.*, 1990; Evans *et al.*, 1995). Sudaryanto *et al.*, (2005) and Rumengan *et al.*, (2008) reported that in sediments from Indonesia the total butyltins (TBT, DBT and monobutyltin) concentrations ranged from 0.51 to 320 ng/g dw in 1998. The TBT concentrations we found in this study were higher than above reported concentrations in samples from Indonesia. Our data suggest that TBT contamination is still occurring in Indonesia. It is worth noting that the sediments from the shipyards in

ports are periodically discharged into the sea, which could result in widespread contamination in the coastal areas of Indonesia. Butyltins has been identified as a contaminant of potential concern in the marine sediment (Rumengan *et al.*, 2008).

Arai and Harino (2009) reported lower butyltin residue concentrations in Indonesia than have been reported in coastal and harbor areas in other parts of the world including several other developing Asian countries and urbanized industrial nations such as the USA, European countries, Australia and Japan. However, our study, performed in 2011, show that there are high butyltin concentrations in sediments in Indonesia. These results confirmed that, with no current restrictions, there is a high risk of heavy butyltin contamination in the marine of Indonesia.

A high concentration of TBT ( $0.63 \mu\text{g/g ww}$ ) was detected in Hakata port, Japan, which suggests either a fresh input of TBT into the environment or only slow degradation processes in this region. TBT concentrations in sediment in Japan have decreased, but TBT is still detected even though the use of TBT in Japan has been strictly regulated since 1990.

TBT concentrations were lower in sediments from Japan than in sediments from Indonesia. There appears

**Table 1.** Concentrations of DBT and TBT ( $\mu\text{g/g}$  dry weight) in sediments collected around the world  
 \* wet weight; na=no data available; nd=not detected

Location	Area description	Sampling year	DBT	TBT	References
China	Xiamen port	2002	nd	0.077	Huang <i>et al.</i> , 2005
Hawaii	Pearl Harbor	Published 1989	0.004–0.367	0.004–2.83	Kram <i>et al.</i> , 1989
Hong kong	Harbour	2002	na	<0.0001–0.56	Cheung <i>et al.</i> , 2003
India	Ship building, ship repair, dry dock and small jetties	2000–2002	nd–0.469; nd–0.131	0.016–16.816; 0.0045–1.193	Bhosle <i>et al.</i> , 2006
Indonesia	Port, urban and industrial area	1998	<0.0027–0.087	0.00051–0.19	Sudaryanto <i>et al.</i> , 2005
Indonesia	Manado, Bitung port, Gangga Island & Jakarta Bay	2004	0.0019–0.012	0.0007–0.35	Harino <i>et al.</i> , 2012
Indonesia	Port & shipyard	2011	nd–1.74	nd–4.25	This study*
Japan	Otsuchi Bay	1996	na	0.01–0.64	Harino <i>et al.</i> , 1998
Japan	Port of Osaka	1995–1996	na	0.01–2.1	Harino <i>et al.</i> , 1998
Japan	Maizuru Bay	2003	0.0023–0.023	0.0012–0.019	Ohji <i>et al.</i> , 2007
Japan	Hiroshima Bay	2003–2004	0.0013–0.14	0.0012–0.17	Onduka <i>et al.</i> , 2008
Japan	Hakata Port,	1998	na	1.1	Inoue <i>et al.</i> , 2002
Japan	Port & shipyard	2010	nd–0.214	nd–0.631	This study*
Korea	Enclosed bay	1995	0.01–0.573	0.004–0.382	Shim <i>et al.</i> , 1999
Malaysia	Area of urban, aquaculture, industrial, shipping line and Port	1997–1998	0.0038–0.31	0.0028–1.1	Sudaryanto <i>et al.</i> , 2004
Thailand	Coastal area	1995	0.0002–1.9	0.004–4.5	Kan–Atireklap <i>et al.</i> , 1997
Thailand	Industrial area	2004	0.001–0.368	0.002–1.246	Harino <i>et al.</i> , 2006
Vietnam	North & central coastal areas	2002	0.00064 –0.0046	0.00089–0.034	Midorikawa <i>et al.</i> , 2004

to be an increasing TBT concentration trend in sediment in Indonesia from 1998 (Sudaryanto *et al.*, 2005; Rumengan *et al.*, 2008; Harino *et al.*, 2012) to 2011 (our results). In contrast, a decreasing trend was observed in TBT sediment concentrations from 1998 (Inoue *et al.*, 2006) to 2010 (our results) in northern Kyushu, Japan. The difference in TBT and DBT concentrations and trends is likely to be related to the fact that these compounds have been banned in Japan, whereas no regulations on their use have been implemented in Indonesia.

TBT contamination trends are shown in Table 1, in which the concentrations of butyltin in various parts of the world are compared with the data from our study. High concentrations of TBT were found in a coastal area in Malaysia (1.1  $\mu\text{g/g}$  dw; Sudaryanto *et al.*, 2004), in an industrial area where many tankers and cargo ship in Thailand (1.25  $\mu\text{g/g}$  dw) (Harino *et al.*, 2006) and in Kochi (16.82  $\mu\text{g/g}$  dw) and Mumbai (1.19  $\mu\text{g/g}$  dw) harbors in India (Bhosle *et al.*, 2006). Harbors area, of course, the busiest part of port areas, and often include ship building, ship repair, and dry dock facilities, together with small jetties for berthing fishing boats. The highest TBT concentration found in sediment (12.4  $\mu\text{g/g}$  dw) was associated with shipyard activity (Page *et al.*, 1995). In our study, the highest TBT concentration (4.25  $\mu\text{g/g}$  ww) was also recorded in sediment collected from a shipyards area. This trend is consistent with the sediments being highly affected by shipping activities, and shipyards being mostly responsible for the TBT and DBT contamination in the ports.

High TBT concentrations were found in specific areas in Indonesia and the northern Kyushu coastline, particu-

larly in sediments near shipyard. Contaminated wastes and wastewater from the ports are important potential sources of TBT pollution. There are many potential present and future sources of TBT contamination to the marine environment. Our results suggest that shipyards areas in ports may be major sources. Adverse effects on non-target biota are possible because sediment re-suspension can release TBT into the water column (Harris *et al.*, 1991; Page *et al.*, 1995).

## CONCLUSIONS

High TBT concentrations were found in sediment samples collected near and in front of ports in Japan and Indonesia. Our results support the supposition that shipyard activities are major sources of TBT pollution in Indonesia and Japan, even though TBT usage has been regulated in Japan since 1990.

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