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Contamination of Tributyltin in Sediment from Four Bays in the Southeastern Part of Korea

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Spatial and vertical variations of butyltins (BTs) were investigated in polluted sediments from Jinhae, Busan, Ulsan and Yeongil Bays located in the southeastern part of Korea. Tributyltin (TBT) as a dominant species was detected in 18 of the 20 surficial sediments, and TBT levels were lower than for those in heavily polluted areas worldwide, ranging from 12 to 766 ng Sn/g dry wt. Distribution of TBT levels among bays was not significantly different ($p=0.286$, ANOVA). The horizontal distributions observed at 20 sites suggests that TBT concentrations are still high in polluted sites around harbors and industrial complexes. The sedimentary records revealed that TBT inputs began to increase in the active industrial periods in Korea and reached a peak in the early 1990s in Jinhae Bay, in the mid 1980s in Ulsan Bay and in the late 1970s in Yeongil Bay except for Busan Bay which had a homogenous profile. In paricular, TBT at the surface sediments of four core samples still had high levels. From the horizontal and vertical results, it could be assumed that TBT inputs in the sediments were not reduced significantly compared to past levels. The annual accumulation rates estimated using the sedimentation rates and the concentration of surficial sediments exhibited that the accumulation rate of Ulsan Bay was greater than the rates for Jinhae and Yeongil Bays.

Keywords: Spatial and vertical variation, Sediments, Tributyltin (TBT)

1. Introduction

Tributyltin (TBT) is a very toxic molecule that has been used in marine antifouling paints. The problem with TBT and its cause was first recognized in France[1]. This biocide agent is known to cause deleterious effects in non-target organisms. The two most pronounced effects studied were shell thickening in oysters and imposex in the common dogwelk[1~4]. Therefore, many countries have restricted their use due to evidence of adverse effects on marine life. The Marine Environmental Protection Committee (MEPC) of the International Maritime Organization (IMO) requires that TBT-based antifoulants be totally banned from the year 2003 onwards[5]. Even though TBT concentrations in water from Europe and the US have significantly declined since the regulation of TBT based antifoulants, recent surveys show that TBT concentrations in sediment over time are relatively constant[6~8]. The presence of TBT on sediments makes them available to sediment-feeding organisms. Another risk for the aquatic system is the possible contamination from suspended sediments[9~12]. In 1999, a ban on the use of TBT based paints on fisheries facilities and small vessels was first introduced in Korea. However, high concentrations of TBT still exist, especially in the sediment of harbors and shipyards in Korea due to late regulation, as well as to strong persistence of the sediment-associated TBT[13,14]. In addition to TBT, other organotin compounds such as monobutyltin (MBT), dibutyltin (DBT) and phenyltins have been detected in the aquatic environment. Jinhae, Busan, Ulsan and Yeongil

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Bays located in the southeastern part of Korea are famous as highly polluted regions. Those bays surrounded by industrial cities with a high population density and active industrial ports have strong potential for contamination of TBT. Therefore, it is very important to understand the pollution situation in those areas for the preparation of regulation and management policy on the organic micro-pollutants. As part of such an effort, this study was designed to examine the spatial distribution of a range of butyltin compounds (BTs) in sediments from the industrialized southeastern part of Korea and to explore the input history of these organic contaminants using an interpretation of the sedimentary record.

2. Materials and Methods

2.1. Sampling sites

Jinhae Bay (J1-J5), Busan Bay (B1-B5), Ulsan Bay (U1-U5), and Yeongil Bay (Y1-Y5) in the southeastern part of Korea have been subject to pollution by the industrial wastewaters from a large number of engineering works and sewages generated in industrial cities such as Masan, Busan, Ulsan and Pohang (Fig. 1). Among 20 sampling stations, stations J1, B1, B2, B3 and U1 are located close to big harbors: Masan Harbour, adjacent to J1, has played a pivotal role as the commercial port of the Masan export free area since 1970. Busan Harbor, near B1, B2 and B3, is the largest harbor in Korea with a long history. Ulsan Harbour, close to U1, is also just as famous for its large scale. Station J5 has small wharves for fishing boats. This area is very popular for oyster and mussel cultures. Hyundai and Samsung shipyards worldwide are located in the vicinity of stations U2 and J4, respectively. Station J2 exists in a site around a naval military base. Many larger industrial factories are located very adjacent to stations Y1, Y2, Y3 and U5. Pohang city is highly industrialized, especially because of its steel works, while Onsan city because of petroleum,

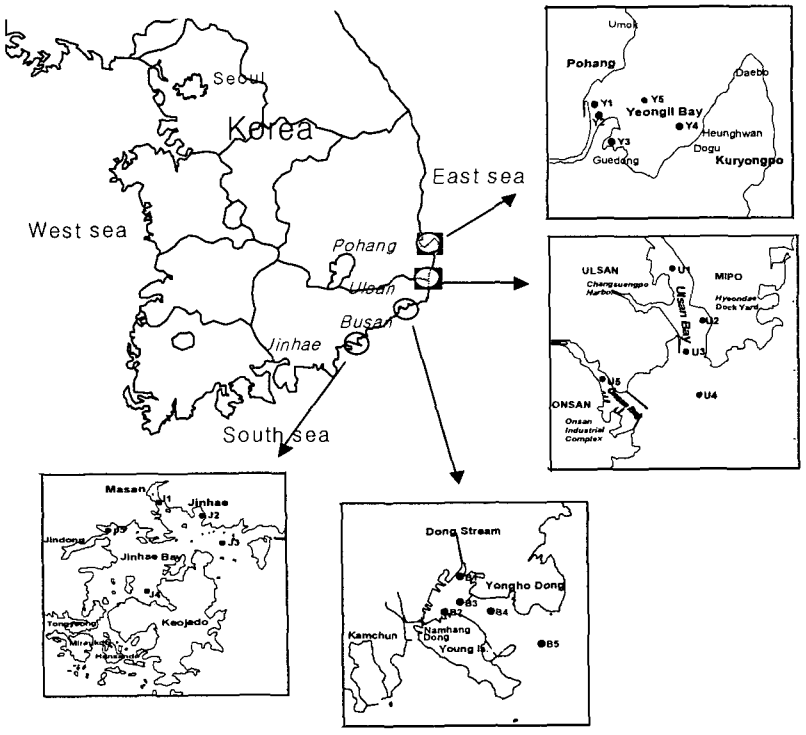

Fig. 1. Sampling sites of Jinhae, Busan, Ulsan and Yeongil Bays in the southeastern part of Korea.

petrochemical and chemical industries. Stations away from shore are J3, B4, B5, U3, U4, Y4 and Y5.

2.2. Sampling and Analysis

Surface sediment samples from the top 0-5 cm were obtained from 20 sites by box core in November, 2000. Sediment cores were also collected by a diver using an acrylic tube (8 cm diameter and 150 cm long) at stations J4 and U2 in September, 1999, and stations B1 and Y2 in September, 2000 (Fig. 1). Care was taken to obtain core samples so as not to disturb floccules existing above the surface sediment. After collection, the samples were freeze-dried and then sieved at 2 mm. Samples were stored at -20℃ until extraction. Separate samples were also taken for organic carbon determination. Sediments (2-5 g) were extracted with 0.1% tropolone-methylene chloride in a 50 mL centrifuge tube. Tripropyltin chloride was spiked before extraction as a surrogate standard. A mixture of 3.3% tetrabutylammonium hydrogensulphate and 16% sodium sulfide was added to the extraction vessel to desulfurize the extract. The total organic extract was reduced to a small volume using a rotary evaporator and then purified on silica microcolumn after propylation with a Grignard reagent (n-propylmagnesium bromide). Finally, the internal standard (tetrabutyltin) was added in concentrated eluates to calculate the recovery. Butyltin quantification was conducted on a Hewlett-Packard 6890 gas chromatograph (GC) equipped with a flame photometric detector with a 610 nm cut-off interference filter. A capillary column HP 5 (5% phenyl methyl siloxane, 30 m × 0.25 mm internal diameter × 0.25 μm film thickness) was used. The temperature program was as follows: initially at 80℃, 80-160℃ at 15℃/min, 160-200℃ at 5℃/min, and isothermal 220℃ for 3 min. The injection port temperature was set at 220℃. Quality assurance and quality control procedure included internal standards, procedural blanks and the analysis of a reference material (PACS-2, NRC, Canada). Extraction recovery was 96% for TBT, 96% for DBT and 93% for MBT. Detection limit of each butyltin compound was about 3 ng Sn/g dry. Organic carbon (OC) was measured by a CHN analyzer (Perkin Elmer 2400) and particle size was determined using standard wet sieving techniques to separate into the two fractions, differentiating the clay-silt component (<63 μm in diameter) from the larger particles. Sedimentation rates using ^{210}Pb profiles and a ^{137}Cs-time marker were determined.

3. Results and Discussion

3.1. Spatial distribution

The concentrations of butyltin species, OC and particle size contents are shown in Table 1. TBT was detected in 18 of the 20 sediment samples. The concentrations of TBT were in the range of 12 ~ 766 ng Sn/g dry wt. The high TBT levels were detected at stations B3 and U1, close to harbors and a shipyard with more than 700 ng Sn/g dry wt, and followed by stations Y1, B2 and U2 with more than 300 ng Sn/g dry wt. These results were lower than the levels found in heavily polluted areas

Table 1. Butyltins concentrations (ng Sn/g dry wt), organic carbon (OC) contents (%) and grain size contents (<63 μm, %) in sediments from the southeastern coastal areas of Korea

		MBT	DBT	TBT	∑BTs	OC	<63 μm	Normalized TBT
Jinhae	J1	104	79	114	298	2.56	98	4456
Bay	J2	49	23	16	88	2.21	96	733
	J3	nd	8	30	38	1.05	77	2824
	J4	101	76	105	282	3.33	99	3145
	J5	76	33	43	152	2.59	98	1666
Busan	B1	167	222	255	643	2.23	95	11421
Bay	B2	215	180	345	740	2.32	81	14861
	B3	331	404	766	1502	2.34	96	32754
	B4	77	47	78	202	1.76	80	4414
	B5	45	22	18	85	1.37	99	1312
Ulsan	U1	506	283	706	1495	1.26	85	55994
Bay	U2	1146	283	343	1772	1.11	87	30891
	U3	225	103	196	524	1.56	93	12582
	U4	40	23	18	81	1.18	88	1563
	U5	32	14	14	61	4.64	38	301
Yeongil	Y1	83	96	370	549	1.35	72	27422
Bay	Y2	14	nd	12	26	3.68	81	331
	Y3	47	23	52	122	1.08	92	4791
	Y4	nd	10	nd	10	1.2	86	nd
	Y5	39	nd	nd	39	1.02	79	nd

nd=not detected

worldwide. Kan-Atreklap *et al.* found that the TBT concentrations in sediments along Thailand's coasts were up to 1,837 ng Sn/g dry wt[15]. Page *et al.* reported that TBT concentrations in sediments from Boothbay Harbor, Maine, were 5,061 ± 1,294 ng Sn/g dry wt[6]. According to Gabrielides *et al.*, TBT concentrations ranged up to 1,538 ng Sn/g dry wt in the Mediterranean[16]. This degree of enrichment may pose a threat to aquatic communities if TBT becomes available following sediment disturbance, particularly to benthic organisms[17]. The distribution of TBT reflected boat usage patterns within the water bodies, with enrichment occurring at marina sites, boatyards and mooring localities[18]. Slightly elevated TBT levels at stations adjacent to harbors and shipyards indicated that antifouling paints were a major source of these compounds in the aquatic environments of southeastern coastal areas. However, it was somewhat surprising that station B3 exhibited higher concentrations of TBT compared with stations B1 or B2 located at inner sites of Busan Bay. This is likely to be related to physical transportation factors such as flow patterns and/or ship traffic. The distributions of TBT concentrations among the four areas were not significantly different even though relatively high TBT concentrations were detected in sediments from Busan and Ulsan Bays ($p=0.286$, ANOVA). On the other hand, TBT is a well-known particulate-associated contaminant, showing high levels in fine sediments[12]. Therefore, particle size and organic carbon contents were measured (Table 1) and statistical comparisons of TBT concentrations among the four bays were performed using carbon-normalized values to reduce the effect of inter-area variations of organic carbon. The results showed a poor correlation between organic carbons and TBT concentrations. This might result from the fact that almost all of the sediments in the bays were predominantly silt-clay (<63 μm). The distributions of the normalized TBT concentrations through a sqrt data transformation also did not show a significant difference among the areas ($p=0.273$, ANOVA). Regulations regarding TBT in Korea were adopted in September 1999. In the comparison of TBT levels between previous studies[13,14,19] and this study, there was no evidence that TBT concentrations decreased in 2000. Japan banned the use of TBT-based ship paints in 1990. Harino *et al.* reported that TBT levels in the sediments in the port of Osaka did not change between 1990 and 1996[7]. The United Kingdom (UK) introduced legislation to control the sale of TBT based paints in 1986. According to Waite *et al.*, changes in concentrations of TBT in sediments showed no clear trend from 1986 to 1989 in spite of the reduction in concentrations of TBT noted in water and bivalves[20]. The problem is that the half-life of TBT in sediments may be a matter of years or even decades[21], and TBT sorption onto sediment can be reversible into the water column[6,18,22]. It means that marine organisms can be affected by the release into the water column of sediment-associated butyltin. Therefore, the periodic monitoring and estimation on the marine ecosystem must be carried out carefully in order to mitigate the environmental impact by development plans such as dredging. The ratios of TBT to the total BTs in sediment accounted for more than 50% at the highly polluted stations B2, B3, U1, U2 and Y1, while they were less than 23% at the unpolluted stations B5 and U3 (Fig. 2). From these results, it could be assumed that the ports still have new inputs of TBT.

3.2. Depth profile

The ^{210}Pb and ^{137}Cs contents in sediment cores are presented in Table 2. The sedimentation rates calculated by ^{210}Pb profiles were 0.42 cm/yr for Jinhae Bay (station J4), 1.32 cm/yr for Ulsan Bay (station U2) and 0.14 cm/yr for Yeongil Bay (station Y2). Busan Bay (station B1) exhibited homogenous ^{210}Pb activities with depth. There was close agreement on sedimentation rates exhibited between ^{210}Pb and ^{137}Cs based methods. Vertical profiles of TBT in the sediment core of four bays in the southeastern part of Korea are shown in Fig. 3. Tributyltin compounds were present in the sediment from the surface layer down to the bottom of the cores sampled throughout the areas. This means that the sampling sites have had continuous influence from pollution sources such as harbors, shipyards or industrial complexes. Down-core trends

of butyltins in Jinhae and Yeongil Bays were characterized by a decrease from the surface horizons to depth. The historical trend of Ulsan Bay was dominated by a distribution of a subsurface maximum with decreased contaminant levels both to the surface and with depth in

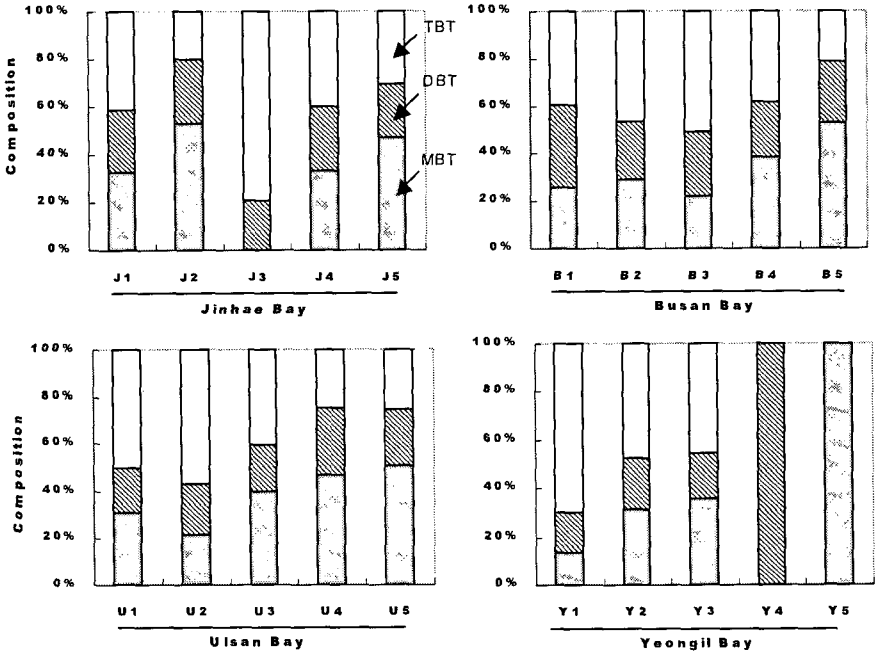

Fig. 2. Contribution of TBT and its degradation products (DBT and MBT) to total butyltin compounds in the surficial sediments at 20 stations from the southeastern part of Korea.

Table 2. Depth profiles of ^{210}Pb, ^{226}Ra and ^{137}Cs activities (dpm/g) in sediment cores from four bays in the southeastern part of Korea

Depth	Jinhae Bay (J4)			Busan Bay (B1)			Ulsan Bay (U2)			Yeongil Bay (Y2)		
(cm)	^{210}Pb	^{226}Ra	^{137}Cs	^{210}Pb	^{226}Ra	^{137}Cs	^{210}Pb	^{226}Ra	^{137}Cs	^{210}Pb	^{226}Ra	^{137}Cs
0-2	6.05±0.44	0.94±0.12	0.29±0.11				10.02±1.27	1.72±0.21	0.3±0.11	7.80±0.94	1.54±0.19	0.17±0.10
2-4	5.98±0.86	0.59±0.17	0.27±0.08	8.25±1.10	1.70±0.22	0.18±0.10				14.34±1.94	3.01±0.35	0.38±0.14
6-8	4.76±0.59	1.17±0.13	0.24±0.07							8.15±1.08	2.31±0.23	0.28±0.13
8-10	4.45±0.52	0.95±0.18	0.28±0.13				9.14±1.25	1.96±0.22	0.18±0.10	4.96±0.91	1.62±0.19	0.20±0.10
10-12	6.30±0.71	0.58±0.22	0.33±0.13	8.01±1.06	1.62±0.20	0.18±0.09				3.52±0.68	1.59±0.13	0.12±0.06
12-14												
14-16	5.95±0.42	1.03±0.14	0.35±0.09				7.14±1.01	2.05±0.16	0.20±0.07			
18-20	3.77±0.75	1.39±0.18	0.10±0.07	8.71±1.16	1.38±0.22	0.39±0.13				3.89±0.75	1.81±0.16	0.16±0.07
20-22	2.95±0.67	1.30±0.17					7.37±1.18	1.88±0.22	0.16±0.10			
26-28	1.52±0.53	1.04±0.14					7.34±0.73	2.21±0.16	0.21±0.07	4.20±0.62	1.98±0.14	0.18±0.05
28-30												
34-36				8.47±1.21	1.73±0.24	0.27±0.14	6.30±0.92	2.00±0.19	0.40±0.08	4.02±0.83	1.84±0.17	
36-38												
42-44							5.05±0.97	2.29±0.21	0.33±0.10			
46-48							3.31±0.40	2.19±0.14	0.38±0.08			
50-52							5.98±0.92	1.64±0.18	0.51±0.10			
58-60							6.13±1.08	2.20±0.25	0.40±0.11			
64-66							5.64±1.13	1.67±0.24	0.30±0.12			

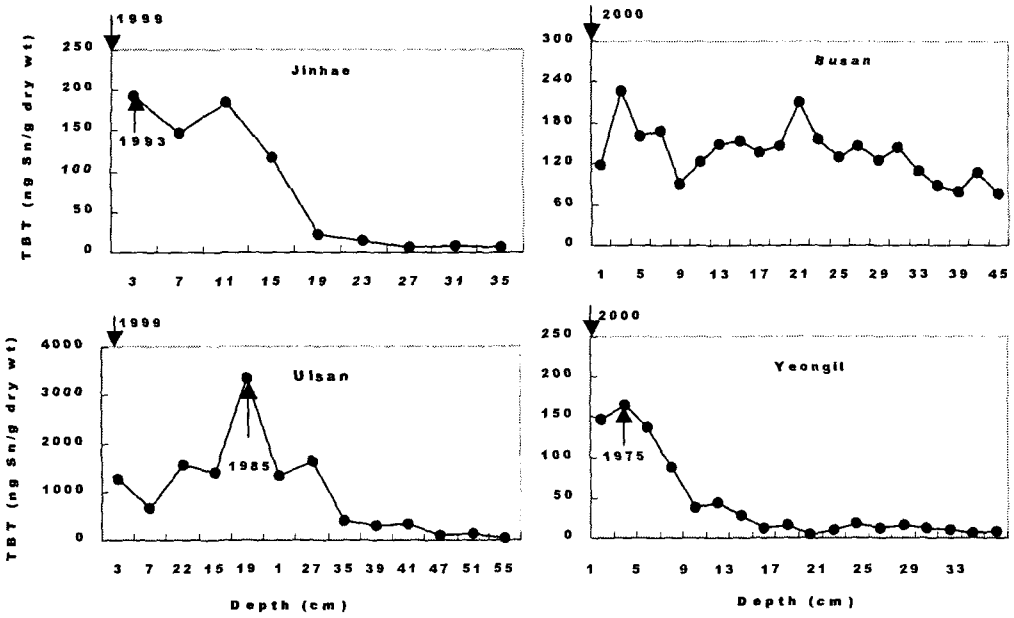

Fig. 3. Depth profiles of TBT concentration in sediment cores from four bays in the southeastern coastal areas.

the core. Maximum concentrations of TBT were detected at depths of 2-4 cm for Jinhae Bay, 2-4 cm for Busan Bay, 18-20 cm for Ulsan Bay and 2-4 cm for Yeongil Bay. These sections of the core correspond approximately to the early 1990s in Jinhae Bay, in the middle 1980s in Ulsan Bay, and in the late 1970s in Yeongil Bay. The pollutant profiles indicate that the present-day situation has not largely improved from the contaminated conditions in the 1970s or 1980s. The use of TBT compounds in marine antifouling paints began between 1959 and 1961. By 1985, an estimated 20-30% of vessels worldwide utilized TBT containing antifouling paint systems[23]. However, TBT concentrations reduced during the 1980s and 1990s due to the replacement of TBT free association coatings by co-polymer paints and regulations introduced by several governments[3]. Korea entered a period of rapid industrial development in the 1960s and major industrial development have progressed in Masan, Busan, Ulsan and Pohang located in the southeastern coastal areas. Therefore, it could be noticed that these historical contaminations were influenced until recent years by the harbors developed around those industrial cities after the onset of industrialization. The Busan core showed a homogenous profile, suggesting the active biological activity of benthic organisms as well as the physical mixing of such things as the transportation of ships. Many authors reported that correlation between butyltin concentrations and depth depends on several factors such as: the variance of introduction rates of butyltin pollutants in time, the differences in sediment composition, hydrodynamical conditions, bioturbation/mixing and sedimentation rates[6,24]. TBT percentage profiles in core sediment samples are presented in Fig 4. The percentages of TBT in sediment samples are an approximate measure of the extent to which the TBT has undergone biodegradation. Aboul Dahab *et al.*[25] observed an increasing contribution of MBT and TBT with depth, and Stan & Seligman[26] reported a direct degradation from TBT to MBT. The contribution of its degradation products (DBT and MBT) to TBT was estimated in sediment core samples from surface to depths corresponding to the 1960s. The organotin compounds have been used as antifouling paints since the 1960s. The contributions of TBT in sediment cores from Jinhae, Ulsan and Yeongil Bays

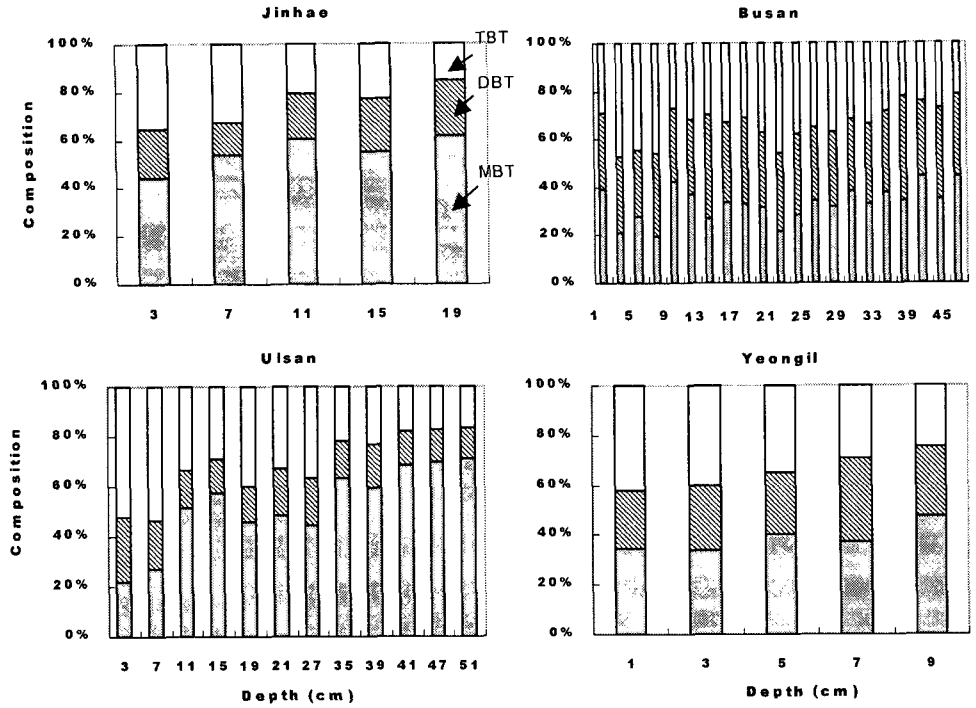

Fig. 4. Contribution of TBT and its degradation products (DBT and MBT) to total butyltin compound in sediment cores from four bays in the southeastern part of Korea.

exhibited a weak decrease with depth due to biodegradation.

3.3. Accumulation rate

The annual accumulation rates into the sediments can be obtained by using the sedimentation rates and the concentration of surficial sediments. The component accumulation rate (ω) is based on the following equation: $\omega = C \cdot SR \cdot (1-\varphi)\rho$

Where C is the sediment concentration, SR is the sedimentation rate, φ is porosity, and ρ is the sediment core density. According to Yang *et al.*[27], the mean sediment density and porosity in sediments with a 50 to 90% moisture content can be assumed as 2.5 g/cm^2 and 0.8, respectively. In this study, the moisture contents ranged between 50% (Y2) and 80% (B1). Therefore, the sediment density of 2.5 g/cm^2 and porosity of 0.8 was applied to estimate the accumulation rates of TBT in the southeastern coastal areas. The estimated TBT accumulation rates into the sediments are shown in Table 3. The accumulation rate of Ulsan Bay was 20 times and 83 times greater than the rates of Jinhae and Yeongil Bays, respectively. Unfortunately, it was quite difficult to depict the geographical variation of BT accumulation rates because of the lack of comparable data so far reported.

Table 3. Sediment accumulation rates from four bays in the southeastern part of Korea

	Sedimentation rate (cm/yr)	TBT (ng Sn/g dry)	Accumulation rate (g/cm^2/yr)
Jinhae Bay	0.42±0.11	193	81±21
Busan Bay		119	
Ulsan Bay	1.32±0.36	1,257	1,659±453
Yeongil Bay	0.14±0.01	146	20±1

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