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Tributyl tin levels for sea water, sediment, and selected marine species in coastal Northland and Auckland, New Zealand

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Abstract Water and sediment samples were collected from the Waitemata Harbour, Opua Inlet, and Tutukaka Harbour, and analysed for tributyl tin (TBT) using a hydride generation atomic absorption method. Specimens of oysters (*Crassostrea gigas*, *Saccostrea glomerata*, *Ostrea heffordi*) and oyster borer, *Lepsiella scobina*, were collected from the Tamaki Estuary and surrounding areas. These specimens were analysed for TBT using graphite furnace atomic absorption, and the oyster shell valves were examined for thickening. Concentrations of TBT in the water column were as high as 0.32 μg TBT-Sn l^{-1}. Marina sediments contained up to 0.240 μg TBT-Sn g^{-1}. Shell thickening was found to occur in *C. gigas* where its incidence was correlated with the TBT body burden. Concentrations up to 2.24 μg TBT-Sn g^{-1} (on a dry weight basis) were observed for *C. gigas*. This survey provides evidence for the toxic influence of TBT on non-target organisms in New Zealand.

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INTRODUCTION

Tributyl tin (TBT) is the active anti-fouling constituent in some anti-fouling paints. Goldberg (1986) stated that TBT compounds are "probably the most toxic compounds ever deliberately introduced by societies into natural waters". The very high toxicity of TBT to certain marine organisms has been demonstrated in several studies. Beaumont & Budd (1984) found a 15-day LC_{50} for *Mytilus edulis* (the common mussel) of 0.02 μg TBT-Sn l^{-1}. Reduced compensation for hypoxia and shell thickening occurs in *Crassostrea gigas* (the Pacific oyster) at 0.004 μg TBT-Sn l^{-1} (Lawler & Aldrich 1987). Studies by U'ren (1983), Gibbs & Bryan (1986), and Waldock & Thain (1983) have shown lethal and sublethal effects at similar concentrations. In the environment, the use of TBT in anti-fouling paints has been linked with deformities to oysters in France (Alzieu et al. 1982) and the decline of gastropod populations around south-west England (Bryan et al. 1986). Preliminary studies have suggested that similar effects may be occurring in New Zealand with respect to imposex in *Lepsiella scobina*, the oyster borer (P. Smith, pers. comm.). Imposex refers to the development of male sexual characteristics in females and may cause infertility. Furthermore, instances of shell thickening in *C. gigas* and *Saccostrea glomerata* (the native rock oyster) in New Zealand have been noted, although the tissue content of TBT had not been determined (Smith & Curtin 1986).

The purpose of this study was to assess the impact of tributyl tin on the marine environment around Auckland and Northland, New Zealand. The presence of TBT was determined in various samples of sea water, sediment, and selected marine organisms. This survey of TBT levels was performed in conjunction with an investigation of shell thickening in three species of oyster: *C. gigas*, *S. glomerata*, and *Ostrea heffordi* (the mud oyster).

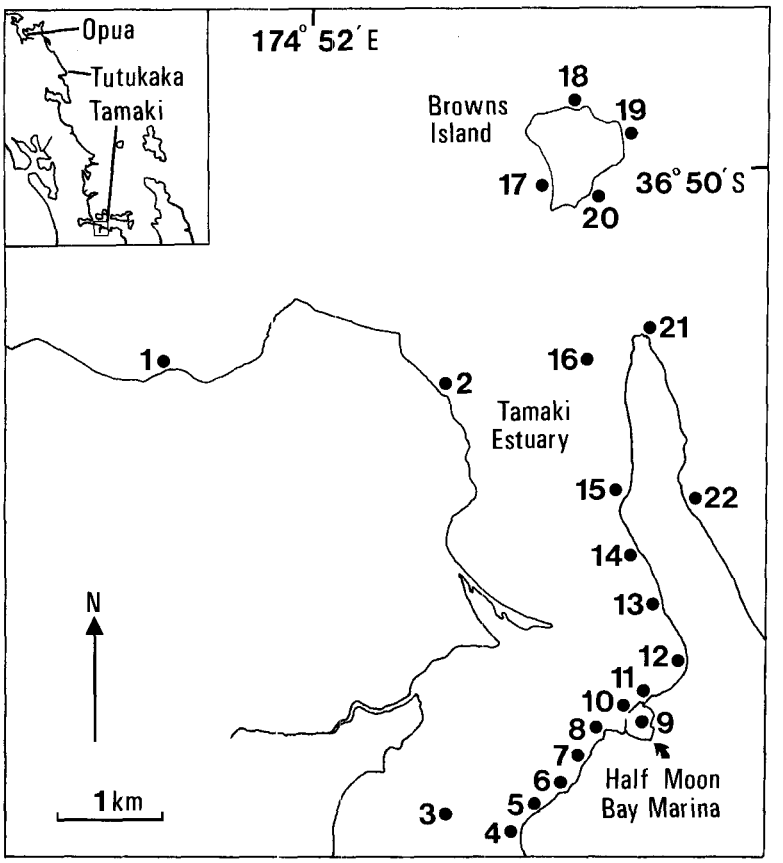

Fig. 1 Sampling stations within the Tamaki Estuary and immediately adjacent marine locations.

MATERIALS AND METHODS

Samples were collected from 4 locations. Opua Inlet (35°19′S, 174°07′E) lies in the Bay of Islands and contains a mooring area with about 300 boats on swing moorings. Tutukaka (35°37′S, 174°32′E) is situated on the coast north of Whangarei and has a marina used by about 100 boats. Westhaven Marina (36°50′S, 174°45′E) lies within the Waitemata Harbour and holds 1500 boats. The Tamaki Estuary (36°50′S, 174°52′E) is situated on the edge of the Waitemata Harbour and contains a 500 berth marina at Half Moon Bay as well as many swing moorings. Samples of sea water and sediment were collected from each of the 4 locations. Sampling stations in the Tamaki Estuary are shown in Fig. 1; all other stations are given in King (1988). Stations were chosen in an attempt to obtain a wide range of tributyl tin concentrations. Accordingly, samples were collected from within marinas (high contamination suspected) to open waters up to 10 km from the nearest marina (low contamination suspected). In addition to sea water and sediment samples, oyster specimens (*C. gigas*, *S. glomerata*, and *O. heffordi*) and oyster borer (*L. scobina*) were collected from the Tamaki Estuary and adjacent marine locations.

Sea water samples were collected from a depth of 0.5 m directly into 125 ml polyethylene bottles and acidified to 0.1 mol l^{-1} HCl. Sediment and biological samples were placed in polyethylene self-sealing bags. All samples were frozen as soon as possible after collection.

The concentrations of TBT in samples of sea water were determined using a hydride generation atomic absorption system based on that used by Hodge et al. (1979) and described in King (1988). This analytical system separates TBT from the other tin species present in an environmental sample, including the products of TBT degradation. The detection limit (3 standard deviations or σ of the blank) for this system was 0.005 µg TBT-Sn l^{-1} for a 100 ml sea water sample. However, sea water samples were not considered quantifiable unless they exceeded 0.016 µg TBT-Sn l^{-1} (10σ).

Dried sediment samples (about 2 g) were treated with 10 ml 1 mol l^{-1} HCl. Following a 2-h digestion period, the sediment-acid mixture was quantitatively transferred to the reaction vessel of the hydride generator. TBT was analysed as noted above for sea water samples. The detection limit for a 2 g sediment sample was 0.001 µg TBT-Sn g^{-1}. All sediment concentrations have been expressed on a dry weight basis. TBT concentrations in biological material were measured using a method based on that of Short & Thrower (1986). The exact method used is described in King (1988) and involves extracting the TBT into hexane and determining the tin level of the extract by graphite furnace atomic absorption. Because some dibutyl tin is carried through in the extraction, the method is not specific for TBT. The detection limit (3σ) for oyster tissue was 0.013 µg TBT-Sn g^{-1} (dry weight).

To relate oyster shell deformity to TBT body burden, it was necessary to quantify the degree of deformity. A shell thickness index was used based on the number of cavities present within the valves of each oyster.

RESULTS AND DISCUSSION

The content of TBT found in sea water samples is given in Table 1. Concentrations range from below the detection limit (0.005 µg TBT-Sn l^{-1}) to 0.32 µg TBT-Sn l^{-1}. Out of 34 samples analysed, TBT concentrations exceeded the limit of quantification (0.016 µg l^{-1}) in only 10 cases. The highest levels were found in marinas although some well-flushed mooring areas contained no detectable TBT, as observed at Opua Inlet. The only other data from New Zealand are those of Smith & Curtin (1986) who measured TBT levels at Westhaven Marina (Auckland), Port Nicholson and Evans Bay (Wellington), Picton, and Havelock. Concentrations of total organic tin ranged from 0.04 to 0.78 µg TBT-Sn l^{-1}.

Numerous overseas studies are available for comparison. In general, TBT concentrations are highly variable but tend to be greatest for waters within marinas. In south-west England, Cleary & Stebbing (1985) found a maximum level of 0.18 µg TBT-Sn l^{-1}. Waldock & Miller (1983) measured 0.62 µg TBT-Sn l^{-1} in the Essex marina in south-east England. In Arcachon Bay (France) where oyster deformities were first observed, Alzieu et al. (1986) have measured TBT levels up to 0.18 µg TBT-Sn l^{-1}. Studies in North America have recorded maximum TBT levels of 0.34 µg TBT-Sn l^{-1} in San Diego Bay (Valkirs et al. 1987), and 0.43 µg TBT-Sn l^{-1} in Chesapeake Bay (Hall et al. 1987). Thus TBT concentrations in New Zealand waters are

Table 1 Tributyl tin concentrations in sea water. n.d., < 0.016 µg TBT-Sn l^{-1}

Location	Station	Date	TBT (µg Sn l^{-1})	Comments
Westhaven	1	3/8/87	n.d.	
	2	12/8/87	n.d.	
	3	12/8/87	n.d.	
	4	12/8/87	0.288	
	5	12/8/87	0.110	
	6	12/8/87	0.058	
	7	3/8/87	0.032	
	7	3/8/87	0.062	Surface sample
	8	3/8/87	n.d.	
	9	12/8/87	n.d.	
	9	12/8/87	n.d.	Surface sample
Mission Bay	–	3/8/87	n.d.	
Okahu Bay	–	3/8/87	n.d.	
Tamaki River	7	18/11/87	0.018	
	8	18/11/87	0.016	
(Half Moon Bay)	9	3/8/87	0.110	Average of 2 measurements
	9	18/11/87	0.321	
	12	18/11/87	0.069	
	14	18/11/87	n.d.	
Opua			n.d.	10 sites examined
Tutukaka			n.d.	5 sites examined

comparable to values observed overseas. However, it should be noted in particular that the levels of TBT at Half Moon Bay Marina in the Tamaki Estuary and Westhaven Marina exceed the highest concentrations measured in Arcachon Bay, where oyster deformities occurred over a large area.

The concentration of TBT in the sediments is given in Table 2. In general, the level of TBT in the sediment exceeded that of the overlying waters. Because of this, and the lower detection limit for TBT in the sediment, TBT was found in sediment samples further from the marina sources than TBT in the water column. For instance, TBT was detected in sediments in the Tamaki Estuary 3.5 km seaward of the marina. TBT levels were found to be elevated in areas where high particle settlement rates were suspected to exist (e.g., 500 m down stream of the Half Moon Bay Marina), suggesting that some TBT may be carried in the water column bound to sediment particles.

The TBT content of sediments varies widely. The highest values observed, namely 0.240 and 0.157 μg TBT-Sn g^{-1} (expressed as Sn in dry sediment), were found in and down stream of the Half Moon Bay Marina in the Tamaki Estuary. No TBT was detected (i.e., < 1 ng g^{-1}) in 5 out of 35 sites sampled. Such results are similar to those of Maguire (1984). In examining a number of freshwater sites in Ontario, sediments in Toronto Harbour were the most contaminated having 0.24 μg TBT-Sn g^{-1}

Table 2 Tributyl tin concentrations in sediment. Concentrations are μg Sn g^{-1} dry weight; n.d., < 0.001 μg TBT-Sn g^{-1} for 2 g (dry weight) sediment.

Location	Station	Date	TBT-Sn (μg g^{-1})	Comments
Tamaki Estuary	2	7/12/87	0.001	
	3	4/11/87	0.001	
	5	4/11/87	0.001	
	6	4/11/87	0.001	
	9	7/12/87	0.240	Half Moon Bay Marina
	11	4/11/87	0.157	500 m downstream
	12	18/11/87	0.004	Coarse sand
	15	4/11/87	0.003	
	16	4/11/87	0.001	
	21	7/12/87	n.d.	Sand
	22	4/11/87	0.002	
	23	7/12/87	0.005	Maraetai
Opua	1	15/9/87	n.d.	
	2	15/9/87	n.d.	In mooring area
	3	15/9/87	0.001	In mooring area
	4	15/9/87	0.001	In mooring area
	5	15/9/87	0.002	In mooring area
	6	15/9/87	n.d.	In mooring area
	8	15/9/87	0.001	
	9	15/9/87	0.002	
	10	15/9/87	0.002	
	11	15/9/87	0.001	
Tutukaka	1	16/9/87	0.011	Marina
	2	16/9/87	0.008	Marina
	3	16/9/87	0.010	Marina
	4	16/9/87	0.007	Marina
	5	16/9/87	0.012	Marina
	6	16/9/87	0.001	Downstream
	7	16/9/87	0.001	Downstream
	8	16/9/87	n.d.	Downstream
	9	16/9/87	0.002	Downstream
	10	16/9/87	0.003	Downstream
	11	16/9/87	0.001	Downstream
	12	16/9/87	0.001	Downstream
	13	16/9/87	0.003	Opposite Marina

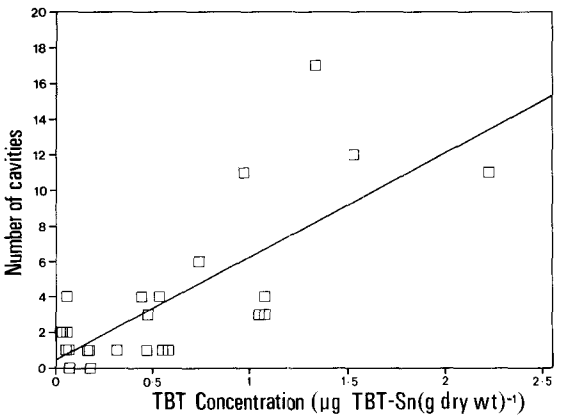

Fig. 2 The number of cavities in individual *Crassostrea gigas* as a function of the body burden of tributyl tin. The solid line represents the best fit by the linear least squares method with a regression coefficient of 0.76.

(expressed here as Sn in dry sediment). In a subsequent survey of 265 locations across Canada, Maguire et al. (1986) found levels as high as 10.8 µg TBT-Sn g^{-1} in the marine sediments of Vancouver Harbour. Much lower concentrations, in the range 0.012–0.044 µg TBT-Sn g^{-1}, were measured for 6 coastal locations in New Hampshire (Randall et al., 1986). Finally, Hattori et al. (1984) examined a single riverine sediment from Osaka, Japan, and found 0.41 µg TBT-Sn g^{-1} sediment. Generally the TBT content in New Zealand sediments is not excessive by international standards. However, elevated levels in the Tamaki Estuary substantiate the trends observed overseas. Sedimentary tributyl tin is assumed to be derived from TBT-based marine anti-fouling paints and as such, marinas can act as significant sources of TBT in the marine environment.

TBT levels were measured in 4 species of marine mollusc: *Crassostrea gigas*, *Saccostrea glomerata*, *Ostrea heffordi*, and *Lepsiella scobina*. TBT was detected in all organisms analysed. Results, averaged by station, are given in Table 3. For all 4 species, the TBT body burden exceeded the TBT level in the surrounding waters by 3 to 4 orders of magnitude. This probably results from passive equilibrium processes rather than active uptake, as the octanol/water partition coefficient for TBT is around 5000 (Laughlin et al. 1986). Overseas data for comparison are available only for *C. gigas*. Alzieu et al. (1986)

Table 3 Tributyl tin concentrations in biological material collected in the Tamaki Estuary grouped by station and species. Detection limit is 0.01 µg TBT-Sn g^{-1} dry weight.

			TBT body burden (µg TBT-Sn g^{-1})		
Species	Station	*N*	Minimum	Maximum	Mean
Crassostrea gigas	1	1	–	–	0.160
	2	2	0.065	0.075	0.070
	4	1	–	–	1.09
	5	2	0.447	0.747	0.597
	6	1	–	–	0.540
	11	6	0.981	2.24	1.38
	13	1	–	–	0.320
	15	2	0.475	0.483	0.479
	16	2	0.175	0.587	0.381
	19	1	–	–	0.555
	21	4	0.028	0.064	0.049
	22	1	–	–	0.182
	Maraetai	1	–	–	0.033
Saccostrea glomerata	11	2	0.218	0.715	0.467
	17	1	–	–	0.176
	18	1	–	–	0.102
	20	2	0.106	0.181	0.143
	21	1	–	–	0.189
	Maraetai	1	–	–	0.049
	Islington Bay	2	0.034	0.054	0.044
Ostrea heffordii	10	3	0.750	1.07	0.931
Lepsiella scobina	4	2	0.62	0.74	0.678
	11	4	0.57	1.94	0.952

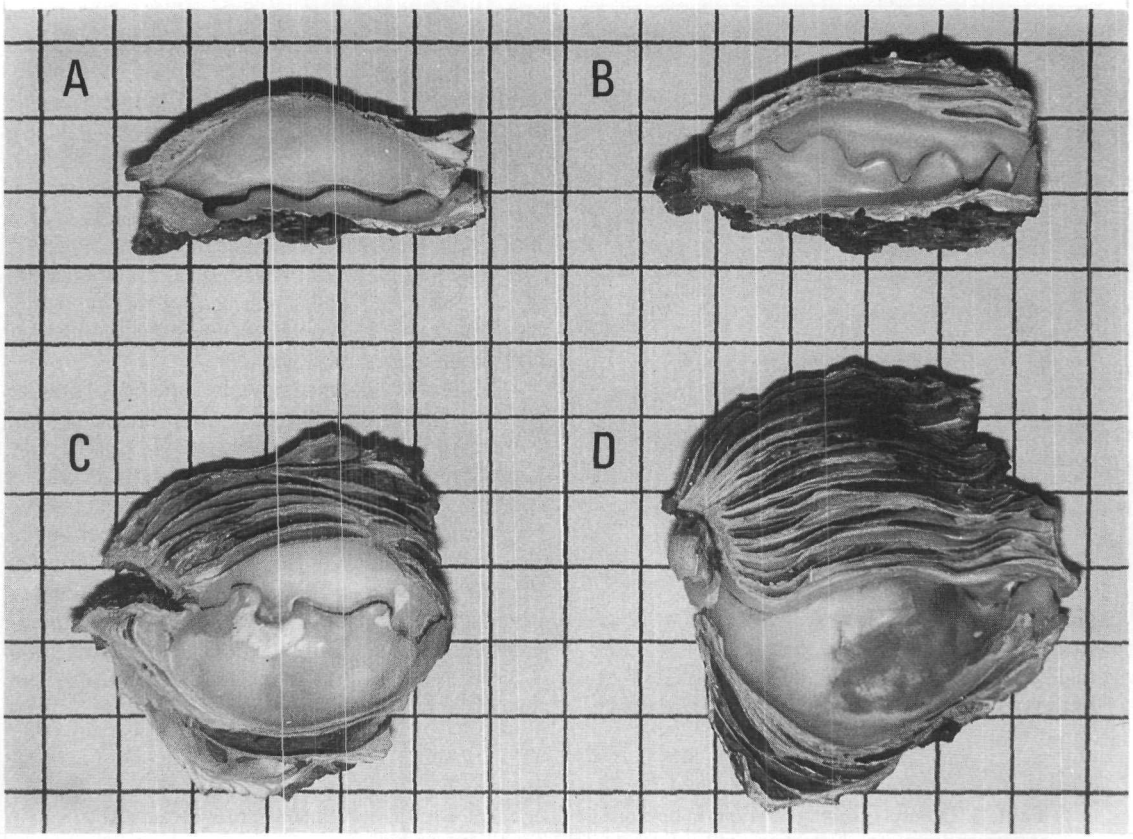

Fig. 3 Specimens of *Crassostrea gigas*, the Pacific oyster, showing shell deformation typical of tributyl tin effects. Shells display thickening and have obvious cavities within the valves. These specimens contain varying amounts of TBT. Expressed as μg TBT-Sn g^{-1} tissue on a dry weight basis, body burdens were: **A**, 0.18; **B**, 1.09; **C**, 1.55; **D**, 0.50. Grid scale 1 cm.

recorded levels of TBT in this species of 0.17–1.65 μg TBT-Sn g^{-1}. This range is similar to that found in the present study (0.028–2.25 μg TBT-Sn g^{-1}).

Of the 3 oyster species examined for shell thickening, only *C. gigas* exhibited evidence of this. Its shell deformation was accompanied by the formation of cavities within the shell valves. Some affected specimens of *C. gigas* were observed at all sites examined within the Tamaki Estuary and adjacent marine sites (see Fig. 1). P. Smith (pers. comm.) has found shell thickening in *S. glomerata* at the Westhaven Marina. Unlike shell thickening seen in *C. gigas*, there were no vacuoles between the shell layers.

The tissue content of TBT in deformed oysters was determined, the first such investigation in New Zealand. The body burden of TBT in *C. gigas* was directly related to shell thickness, and to the presence of cavities within the valves (Fig. 2). Fig. 3 demonstrates the obvious nature of the shell deformation and the influence of varying TBT concentration in the oyster tissue. Thickened *C. gigas* shells have been found at several other sites in the Waitemata Harbour including Kauri Point and Judges Bay. As cavity formation is an obvious manifestation of TBT contamination, surveys of oyster shell deformation would provide a relatively inexpensive means to assess the enviromental impact of TBT.

The level of TBT present in *L. scobina* was not significantly different from that in oysters found at the same sites. This would suggest that *L. scobina* is not accumulating TBT through the ingestion of contaminated food. It is more likely that TBT enters this species by diffusion across the gill surface.

There is some concern that TBT may be adversely affecting the marine environment in New Zealand. The present results tend to justify this concern. The levels of TBT measured in the water column exceed several toxicological thresholds. The maximum concentration recorded in the Westhaven marina was 0.29 µg TBT-Sn l^{-1}, whereas that in the Half Moon Bay Marina was 0.32 µg TBT-Sn l^{-1}. At these concentrations *M. edulis* experiences a significant growth reduction after 144 h (Stromgren & Bongard 1987), 50 % of *Acartia tonosa* (a calanoid copepod) become moribund after 144 h (U'ren 1983), and several algal species experience reduced growth rates (Beaumont & Newman 1984). Of greater concern is that levels in the open waters of the Tamaki Estuary also exceeded several thresholds. At 3 sites in the Tamaki Estuary, TBT concentrations were such that *C. gigas* spat would experience reduced growth over 48 days, reduced compensation for hypoxia over 14 days, and shell thickening over 48 days (Lawler & Aldrich 1987). At 2 sites in the Tamaki Estuary, the 15-day LC_{50} for *M. edulis* larvae (0.02 µg TBT-Sn l^{-1}) is exceeded (Beaumont & Budd 1984).

While not all individuals of *C. gigas* were affected, some degree of shell thickening occurred at every site considered. Tributyl tin was found in all oysters examined within the Tamaki Estuary and environs (see Fig. 1), even where TBT in the water column could not be detected. It is likely that at some sites (e.g., Islington Bay, Maraetai) this TBT originated from low-density anchoring and mooring areas. There is a large number of such areas in the Hauraki Gulf and along the Northland coast, as well as a number of marinas, and so it is probable that many of the shellfish growing in this region will be contaminated with TBT to some degree.

The TBT concentration in the sediments was considerably higher than that measured in the overlying waters. There is no doubt that the sediment acts as a sink for the TBT released from anti-fouling paints. The half-life of TBT in sediments has not been established with any certainty, but may be longer than a year (Seligman et al. 1986). As regards the effect of a ban on the use of TBT, the persistence of TBT in the sediments suggests that sediment TBT concentrations would diminish only slowly and so toxic influences may be exerted for some time. Furthermore, TBT contaminated sediments could be remobilised by storms, currents, and dredging of marinas, mooring areas, and shipping channels. Work is underway at present to examine the fate of TBT in the sediment and the bioavailability of sediment-bound TBT to marine organisms.

CONCLUSIONS

Tributyl tin was measured in a number of areas in the Hauraki Gulf and Northland. Sea water, sediment, and 4 species of marine organisms were investigated. At some sites, TBT levels in the water column and sediments were environmentally significant and exceeded those found in areas overseas where environmental problems have occurred. TBT has a high affinity for lipid-containing tissues and so marine organisms can become contaminated with TBT even when its concentration in the water column is very low. In this study all oysters were found to contain TBT, even when they existed in areas relatively distant (up to 10 km) from large sources of TBT. Similarly shell thickening in *C. gigas* was observed at all sites within the Tamaki Estuary and environs, the degree of deformation being related to TBT body burdens. Such thickening is a manifestation of the toxic influence of TBT on non-target organisms.

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