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INPUTS OF TBT TO THE MARINE ENVIRONMENT FROM SHIPPING ACTIVITY IN THE U.K.

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ABSTRACT

Following legislative measures to ban the use of tributyltin-based antifouling paints on small boats in the U.K., a programme of investigations has been commissioned to examine inputs from shipping. Shipping activity has been divided into three categories for investigation: (a) commercial harbours; (b) anchorages; and (c) dry docks. Significant concentrations of TBT have been measured in harbours and at anchorages, in some cases exceeding 50 ng l^{-1} TBT in the water column in the immediate vicinity of ships. However, the contribution to environmental concentrations of TBT from antifouled yachts caused difficulties in assessment of the inputs from ships alone at many of the sites under investigation. Dry-docking practices have been shown to result in discharge of hazardous concentrations of TBT and such activities are now subject to a voluntary code of practice.

INTRODUCTION

The increasing body of evidence describing the toxic nature of tributyltin (TBT) compounds to aquatic species has led regulatory agencies in several countries to place controls on the use of TBT in antifouling formulations (see 1,2 and 3 for recent reviews). In the U.K., control of the sale of organotin compounds has been exercised since 1986 using the Control of Pollution Act 1974 (4,5,6), and on 1 July 1987, approval for the sale and use of TBT for boats of less than 25 m under the provisions of the Food and Environment Protection Act (1985) (7) was withdrawn, thus banning the use of TBT on yachts and other pleasure craft.

The Department of the Environment (DOE) and Ministry of Agriculture, Fisheries and Food (MAFF) in the U.K., have jointly funded a programme of research to determine the efficacy of control measures. Studies include monitoring the level of environmental contamination by TBT following the ban, examination of recovery of impacted ecosystems and, as described in this paper, measurements of the contribution from shipping activity to the concentrations of TBT found in inshore waters.

These studies focus on the concentrations of TBT at commercial harbours, at anchorages and dry docks.

EXPERIMENTAL PROCEDURE

(a) Commercial harbours

Two sites were selected for study.

(i) Southampton Water (south coast of England) forms a catchment for TBT inputs from a variety of sources including a container ship port, a ferry terminal, a tanker berth at an oil refinery terminal, and pleasure craft marinas. There are several centres of yachting activity in Southampton Water, including the River Hamble where about 5000 yachts are moored in a 5 km stretch of waterway. Surveys of organotin concentrations in Southampton Water were carried out in March and June 1987.

(ii) Harwich harbour, on the east coast of the U.K., has a container ship port at Felixstowe and a ferry terminal at Harwich. Surveys were undertaken in March and June 1987.

(b) Anchorages

Commercial vessel activity is subject to the vagaries of international markets and ships may be moored for a long period of time at one place. Deep and relatively sheltered waters are preferred for such anchorages, which may be some distance away from areas normally used by commercial ships. Three anchorage surveys were carried out, one on the Humber on the east coast of England in February 1987, and two on the Fal in the south-west of England in September 1986 and October 1987.

(c) Dry docks

The purpose of the dry-dock studies was to measure the amount of TBT washed from ships' hulls when vessels were high-pressure hosed before repainting. The input of TBT to harbours near to the dry dock could then be calculated and compared to measured increases in concentrations of TBT in the receiving waters. The information could be used to model the behaviour of TBT released from dry docks, and allow assessment of the potential toxic effects of dry-dock discharges to marine organisms.

Two studies of dry-docking procedure have been carried out:

(i) The washing down of a TBT copolymer formulation from a Leander-class naval frigate was studied in collaboration with the U.K. Ministry of Defence in June 1987 at HM Naval Base, Portsmouth. The frigate was dry docked, washed down with a high-pressure hose systems (17.5 to 21 MPa at 64 $l\ min^{-1}$), and parts of the hull re-sprayed with copolymer paint. Environmental loads of TBT from the washing operation were determined by measurements of TBT concentrations in "hosings" collected from beneath the hull and, by timing the operation, estimates were made of the total amount of TBT removed from the hull over the hosing period.

Details of the construction of the dry dock system and some of the results of the study have already been published (8). Drainage from the dry docks passes to a 500 m x 400 m x 10 m-deep basin which connects to the outer harbour through a series of locks and is very poorly flushed. Measurements of TBT concentrations in this almost enclosed basin before and after the operation could then be compared with calculated quantities discharged.

(ii) Inputs to the environment arising from the washing down of a ship coated with a TBT and copper free-association paint were measured when the MAFF research vessel CLIONE was dry docked at Lowestoft. The study was similar to that for the naval frigate but, in this case, the dry dock discharged directly into the River Waveney estuary. One of the main objectives was to compare the concentrations of TBT in the wash-down water with that generated by the hosing of copolymer paints. Unlike the previous study, the hosing operation began as the dry dock was pumped out. Once the dock was empty, the main part of the hosing operation was carried out, and contaminated water collecting in the bottom of the dock was pumped-out at regular intervals to the estuary. The fate of such a plume of contaminated water was then followed, downstream, from the discharge point.

Analytical methodology

Sampling techniques and the gas chromatographic method used for determination of TBT in water have recently been published (9). Total tin in water was determined by extraction of the acidified water sample by toluene and tropolone and subsequent measurement by Electrothermal Atomic Absorption Spectrophotometry (EAAS) (10,11).

RESULTS

(a) Commercial harbours

Table 1 gives concentrations of organotins measured at various stations within Southampton Water in March and June of 1987. The data for the June survey are summarised in Figure 1. Highest concentrations of TBT were found close to the centres of yachting activity, with over 600 ng l^{-1} being present in the Hamble and one-tenth of this concentration at the mouth of the Itchen, another popular yachting centre. An interesting feature of the survey was the relatively high concentrations of DBT seen at stations remote from boats, accounting for approximately half of the total organotin at the seaward stations.

Water samples taken at the mouth of Southampton Water, near Calshot Castle, in June gave values of 16 ng l^{-1}, and 100 m downstream of the oil refinery at Fawley, 17 ng l^{-1} TBT. Further up the estuary, concentrations of TBT were somewhat higher, with 25 ng l^{-1} at the container port at the top of Southampton Water and 34 ng l^{-1} at Southampton town ferry terminal. The highest concentration observed close to commercial vessels was 78 ng l^{-1}, but this was found within 2 m of an oil tanker at Fawley (Figure 1). A high concentration, 250 ng l^{-1} TBT, was also recorded in March outside a local shipyard where a 3000 t vessel was being hosed down on the foreshore (Table 1).

At Harwich harbour, samples were taken as close as possible to a variety of commercial craft, and concentrations of TBT were also measured from samples taken at reference stations greater than 25 m from the closest vessel. The results in Table 1 show that in March only three, and in August only one, of the samples taken close to vessels were found to contain more than 20 ng l^{-1} TBT. Background values in the harbour appeared to be in the order of a few nanograms per litre in both March and August.

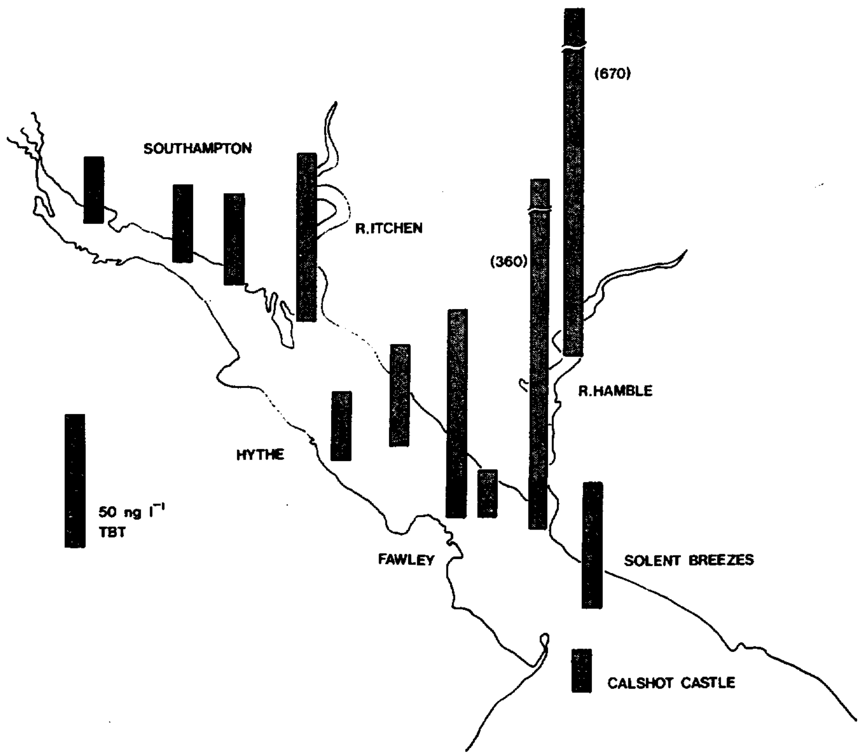

Figure 1. TBT concentrations in Southampton Water

(b) Anchorages

The Humber survey was an opportunistic study, carried out in February 1987 from the MAFF research vessel CLIONE. However, as this vessel is known to be coated with a TBT and copper antifouling paint (see later), the water samples obtained may have been contaminated. Samples were taken over a tidal cycle in the vicinity of 20 vessels of greater than 20,000 t, with the nearest vessel being 700 m away. Throughout the period of sampling, concentrations of TBT did not exceed 2 ng l^{-1} (Table 1).

In the Fal at King Harry Ferry there are usually five to ten commercial vessels at anchor. Depth and distance profiles of TBT concentrations in the vicinity of the moored vessels demonstrated that there was no marked gradient of concentration of TBT away from the ships (Table 1). In the surface waters (1 cm deep) close to one vessel, a concentration of 137 ng l^{-1} of TBT was recorded; however, at 10 cm below the surface, concentrations recorded on an incoming tide were reasonably uniform from 1 m to 1.5 km upstream in the estuary.

Table 1. Concentrations of alkyltin compounds in UK harbours and anchorages

SITE	STATION	DATE	DISTANCE * (a)	DEPTH * (b)	MBT	DBT	TBT	OT
Southampton W	Southampton	18-Mar-87			2	4	42	48
	Cracknore Hard				3	6	2	11
	Husbands Ship Yard				9	76	250	335
	Hythe Pier				2	25	14	41
	Fawley				1	11	17	29
	Calshot Castle	23-Jun-87			2	13	16	31
	Solent Breezes				3	18	46	67
	Hamble Mouth				8	78	362	448
	Hamble Bursledon				8	118	668	794
	Netley Abbey				2	17	38	57
	Itchen Mouth				5	22	63	90
	Town Quay				3	13	34	50
	Top Soton Water				2	10	25	37
	Cracknore Hard				3	22	29	54
	Hythe Pier				7	27	26	60
	Fawley Esso Warwick		2		1	6	78	85
	Fawley Downstrm Esso Warwick		100		3	17	17	37
Harwich	Clervaux	10-Mar-87	5		0 * (c)	0	10	10
	Tor Scandinavia (Ferry)		2		0	5	78	83
Felixstowe	East End of Harbour		35		0	4	1	5
	Aurora		2		0	7	5	12
	Harbour Basin		25		0	2	2	4
	Karen Oltmann		3		0	0	166	166
	Karen Oltmann		50		0	3	2	5
	Mid-Harbour		100		1	2	2	5
	Ile Maurice		4		3	21	3	27
	Dietrich Oldendorff		7		0	9	22	31
	Liesel Essberger		5		0	6	3	9
	Trabant		10		1	8	10	19
	Pegasia		2		0	3	5	8
	Merzario Britannia		2		1	5	3	9
	West End of Harbour		100		2	2	9	13
	Shotley Spit Marker Post		1000		0	7	0	7
Harwich	Laverval	05-Aug-87	2		0	3	12	15
	Bolero		2		0	7	19	26
	Dana Anglia		2		0	6	4	10
	Hamburg		2		0	9	16	25
	Earl William		2		0	3	5	8
Felixstowe	Verrazano Bridge		2		0	0	3	3
	Dorado		2		0	3	7	10
	Strathbrora		2		0	3	3	6

all values expressed as alkyltin cation equivalents in ng l^{-1}. * (a) DISTANCE = distance to nearest moored vessel
* (b) depth of sample = 0.1 m unless otherwise stated. * (c) 0 = less than 1 ng l^{-1}

Table 1. cont.

SITE	STATION	DATE	DISTANCE * (a)	DEPTH * (b)	MBT	DBT	TBT	OT
Felixstowe	Doric Ferry		2		0	2	4	6
	Pegasus		2		1	1	2	4
	Doric Ferry		25		0	2	3	5
	Artus		2		2	6	24	32
	Shotley Spit		2		0	2	8	10
	Nordic Ferry		2		0	0	9	9
	Limassol		2		0	2	5	7
	Eagle Limassol		2		0	1	4	5
	Nordsee Rendsburg		2		0	1	0	1
	Gina S		2		0	3	8	11
	Novamarina		2		1	6	5	12
Humber	Hawke Anchorage	23-Feb-87	700		3	6	2	11
	Hawke Anchorage		700		0	3	1	4
	Hawke Anchorage		700		0	4	0	4
	Hawke Anchorage		700		0	7	0	7
	Hawke Anchorage		700		6	nd	1	7
	Hawke Anchorage		700		0	9	2	11
Fal	Malpas Near Yacht Moorings	02-Sep-86	100		1	7	44	52
	King Harry Ferry		500		1	3	8	12
	King Harry Ferry		200	0.01	1	5	11	17
	King Harry Ferry		200		1	6	17	24
	King Harry Ferry		200	1	1	7	15	23
	King Harry Ferry		200	5	0	3	15	18
	King Harry Ferry		200	10	1	3	6	10
	King Harry Ferry		100		0	4	9	13
	King Harry Ferry		20		0	4	6	10
	Malpas Near Yacht Moorings	20-Oct-87	100		2	6	8	16
	Coombe Creek		100		0	6	4	10
	Earl Granville		1		0	5	35	40
	Earl Harold		1		1	4	10	15
	Earl Godwin		1		1	4	9	14
	Earl Godwin		5		0	4	5	9
	Castor		1		2	12	19	33
	Centaurus		10		0	8	15	23
	King Harry Ferry		0.1	0.01	5	37	137	179
	King Harry Ferry		2		1	4	6	11
	King Harry Ferry		5		1	12	7	20
	King Harry Ferry		10		0	12	3	15
	King Harry Ferry		15		0	2	9	11
	King Harry Ferry		150		0	6	8	14
	King Harry Ferry		400		0	3	7	10
	King Harry Ferry		1000		0	3	5	8
	King Harry Ferry		1500		0	4	3	7

all values expressed as alkyltin cation equivalents in ng l^{-1}. * (a) DISTANCE = distance to nearest moored vessel
* (b) depth of sample = 0.1 m unless otherwise stated. * (c) 0 = less than 1 ng l^{-1}

(c) Dry docks

(i) Copolymer study

Table 2 shows the results of GC analyses of replicate samples of "hosings" from beneath the naval frigate hull. The analytical method employs a contact solvent extraction, and was unlikely to remove all organotin compounds held within larger particles of dried paint. These values have therefore been arbitrarily defined as "readily extractable residues" and may reflect levels of organotin likely to be biologically available in the marine environment in the short term. Larger particles are likely to form a longer-term sink for TBT. Accordingly, to measure the total reservoir of TBT, sub-samples were completely broken down by acid digestion and analysed by EAAS. If all organotin present is assumed to be TBT, mean values by the latter extraction method were about nine times higher than the readily extractable residues.

Table 2. Alkyltin concentrations in replicate samples of washdown water (mg l^{-1})

	MBT	DBT	TBT	Total tin as Sn
FRIGATE				
Aft	0.03	0.03	0.36	3.4
	0.03	0.03	0.33	
	0.01	0.01	0.56	4.8
	0.01	0.01	0.12	
Starboard	0.12	0.36	8.08	14.9
	0.05	0.18	3.08	10.8
	0.00	0.01	0.27	3.2
Port	0.00	0.01	0.61	
	0.02	0.04	0.53	5.1
	0.05	0.18	2.86	
	0.01	0.11	2.49	
	0.00	0.21	3.19	10.0
R.V. CLIONE				
Aft	0.03	0.10	0.32	
	0.07	0.37	0.44	
Starboard	0.06	0.22	0.21	
	0.07	0.21	0.18	
Port	0.06	0.11	0.14	
	0.03	0.07	0.22	
	0.02	0.10	0.23	

Calculation of total input to the marine environment, from time and concentration data, indicates that approximately 100 g of readily extractable TBT was removed from the hull; however, the operation could result in 900 g of TBT being introduced into the environment, a portion of which could eventually become biologically available.

The pre- and post-dry-docking concentrations of organotin in the basin area are shown in Figure 2. The concentrations of TBT in the basin before dry docking ranged from 70 to 240 ng l^{-1}, the highest concentrations being associated with the most poorly flushed section of the basin. There was no clear stratification of TBT concentrations with depth.

The input of 100 g of TBT to the system would have increased the overall concentrations in the basin by approx. 60 ng l^{-1}, assuming complete mixing and no loss to the outer harbour. In fact, concentrations of TBT measured in the basin three days after the hosing operation finished were in the range of 80 to 400 ng l^{-1}.

Depth profiles taken in the outer harbour, two days after hosing, suggested a discharge of TBT from the basin to sub-surface waters of the outer harbour of 50 to 80 ng l^{-1} TBT (Figure 2).

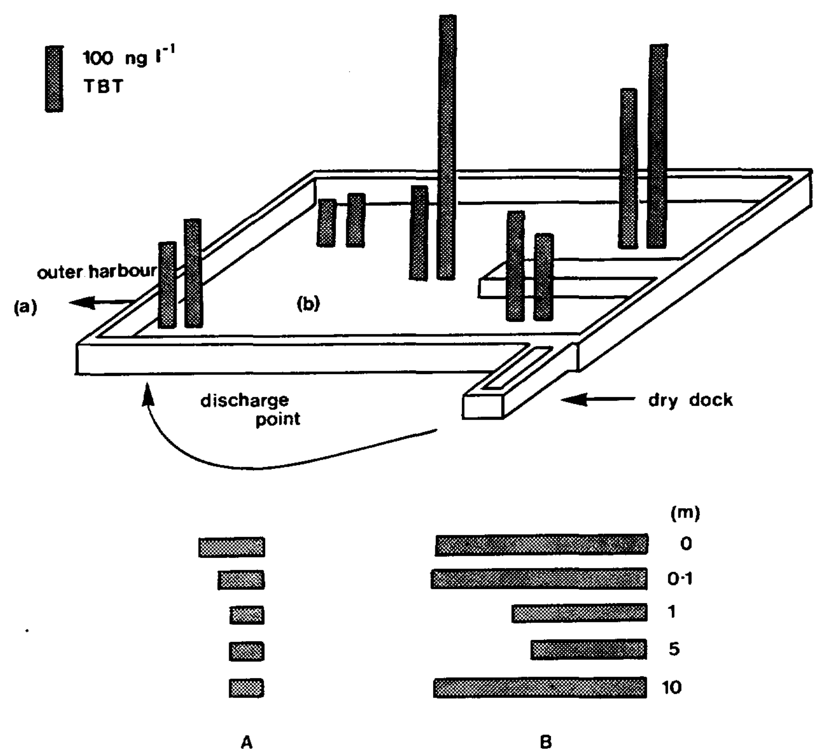

Figure 2. Schematic of Portsmouth naval harbour showing TBT concentrations before (left column) and three days after (right column) washing down of a Leander-class frigate. A and B: distribution of TBT with depth at points (a), and (b) respectively after washing down.

(ii) Free association TBT paint

Concentrations of readily extractable TBT in hosed water beneath RV CLIONE are shown in Table 2. Values are approximately ten-fold lower than those found in the copolymer study. After discharge to the Waveney estuary, highest concentrations of TBT measured were 41,000 ng l^{-1}, representing a dilution of approximately six-fold on discharge. A surface plume of TBT-contaminated freshwater was shown to flow a considerable distance from the dry dock (Table 3). At 300 m the slick was no longer visually identifiable and so it is not clear whether or not the most contaminated water was sampled. It was clear, however, that TBT was present as a surface slick close to the discharge point and concentrations attenuated rapidly with depth.

Table 3. Concentrations of TBT in the Waveney estuary following discharge of wash water from a dry dock.

Distance from discharge point (m)	Depth (m)	Concentration of TBT ($\mu g\ l^{-1}$)
Wash water		250
5	0.1	42
15	0.1	8.7
15	1	0.36
15	5	0.31
50	0.1	0.23
300	0.1	0.22

DISCUSSION

When considering the environmental impact of antifouling paints, an intuitive reaction is to expect that because the immersed hull area of a commercial vessel may be two orders of magnitude greater than that of the average yacht, the input of active ingredient would be proportionally higher. By such reasoning, the 18 container vessels in Harwich harbour should release as much TBT as 1800 yachts. The results of this preliminary survey of TBT concentrations close to commercial vessels suggest that few of the container ships were giving rise to high levels (greater than 20 ng l^{-1}) of TBT in the sub-surface water surrounding the hull. Only two vessels gave rise to more than 50 ng l^{-1} and in both cases TBT could be smelt in the air close to the ships which had recently been painted. Concentrations of TBT in the well-flushed harbour area were close to the limit of detection. Previous experience (9) suggests that a marina containing 1800 yachts in a similar location would give rise to concentrations of TBT at least an order of magnitude higher.

At anchorages, TBT concentrations were also low and in the Fal study most of the moored vessels were heavily fouled. In September 1986, concentrations of TBT close to the anchorage ranged from 6 to 17 ng l^{-1} and were well mixed with depth. However, as a result of moving away from the anchorage upstream towards moored yachts on an ebb tide, the observed concentrations more than doubled. In 1987, concentrations of TBT in sub-surface waters ranged from 3 to 35 ng l^{-1} and only 8 ng l^{-1} was recorded close to yacht moorings on an incoming tide. Assessment of inputs from commercial vessels alone is clearly difficult when yachts painted with TBT share the same waters. The Southampton Water data highlight the problem in that, from this admittedly limited survey, the massive plume of contaminated water from the Hamble appeared to extend at least 3 km to Solent Breezes where small commercial vessels are often anchored. Repeat surveys of Southampton Water are planned once the ban on use of TBT for small vessels is assumed to have taken effect.

The low concentrations of TBT found close to commercial vessels are in accordance with published release-rate figures. Schatzberg (12) suggests that release rates from paints intended for commercial vessels may be five times lower than those for recreational craft (1.0 and 5.0 $\mu g\ cm^{-1}\ day^{-2}$ respectively). Economic considerations dictate that low-release-rate paints are used on commercial vessels in order to reduce the frequency of dry docking (13). Yacht paints, on the other hand, are generally high-release formulations designed to protect moored vessels and may be overpainted several times each year.

Table 4 Inputs of TBT from yacht and ship washdown.

Formulation		Wash water concentration ($mg\ l^{-1}$)	TBT removed from hull (g)	Input ($mg\ m^{-2}$)
Free association	Yacht	1.4	0.4	20
	RV CLIONE	0.25	3.0	10
Copolymer	Yacht	12	3.6	200
	Frigate	2	100	60

The aim of the two assessments of dry-docking procedure recorded here was to determine typical loads of TBT entering the marine environment from hosing down copolymer and free association paints from ships. Hosing-down activities had been identified as giving cause for concern from parallel studies on yachts (9). Comparative results from the hosing of yachts and ships are shown in Table 4. The hosing equipment used in each case was slightly different, and the angle and distance of the water jet to the hull are critical in determining the amount of debris and paint blasted away. Consequently, the estimates necessarily incorporate large margins of error. Nevertheless, the data indicate that washing down of both free association and copolymer painted ships gives rise to lower concentrations of TBT in waste water than from the corresponding formulation designed for yachts. In the case of the free-association paints, this may be due to the yacht paint being less depleted of TBT at the time of hosing and, in the case of copolymers, the formulations used on yachts may be softer than those employed on large vessels. Further studies on hosing down are planned to confirm these observations.

Direct discharge of the hosing-down water to harbours could well be damaging to marine life. The plume of contaminated water arising from the Lowestoft dry dock contained about 40,000 $ng\ l^{-1}$ of TBT. This concentration exceeds the toxic threshold for almost every marine species tested, and is over three orders of magnitude higher than concentrations shown to affect more sensitive species (9). Comparative data for copolymer formulations further suggest that if waste water from hosing a copolymer had been discharged from the Lowestoft dry dock, the environmental concentrations might have been an order of magnitude higher.

In 1985, the U.K. Department of the Environment proposed that for protection of marine species a target concentration of 20 ng l^{-1} TBT should not be exceeded. An environmental quality standard based on present evidence is likely to be set at a concentration some ten times lower (6). These preliminary results suggest that apart from a mixing zone close to commercial vessel hulls, the latest generation of low-release TBT-based formulations for large ships may allow compliance with such strict standards. However, accurate assessment of the relative contribution of ships and yachts to environmental TBT concentrations will not be possible until the ban on use of TBT in the yachting sector is fully effective.

Unregulated dry-dock practices clearly result in the release of large quantities of TBT. In order to prevent environmental problems occurring in the U.K. from these sources, the DOE and shipping representatives have jointly produced a voluntary code of practice for application and removal of antifouling paints, designed to minimise contamination of marine waters (14). It is clear that this code must be followed, preferably for application and removal of all toxic antifouling paint and not only for TBT formulations.

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