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Reductions in TBT Concentrations in UK Estuaries Following Legislation in 1986 and 1987

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ABSTRACT

*In 1986 the Government of the United Kingdom (UK) introduced legislation to control the sale of tributyltin (TBT)-based paints. From 1986 to 1989 monitoring was undertaken, whereby samples of water, sediment, oysters (*Crassostrea gigas*) and mussels (*Mytilus edulis*) were collected from six estuaries and water samples from five marinas/harbours, and analysed for TBT. In 1989 the concentrations of TBT in water, oysters and mussels were generally only one-third to one-quarter of those observed in 1986. The most marked decrease in concentrations of TBT in oysters and mussels occurred in 1987–1988, following the 1987 ban on the use of TBT-based paints on small boats and mariculture equipment. As the concentrations of TBT in oyster tissues has decreased there has been an improvement in oyster growth in terms of both meat production and growth of thin (i.e. normally shaped) shells. Oysters with a normal shell shape can now be grown in five of the six estuaries which were monitored. In contrast to the reduction in concentrations of TBT noted in water and bivalves, changes in concentrations of TBT in sediments showed no clear trend from 1986 to 1989.*

INTRODUCTION

In the early 1970s the Pacific oyster, *Crassostrea gigas*, was introduced into Britain for cultivation. A few years later oyster farmers from estuaries on the east coast of England reported that oysters were growing with abnormally thickened shells. Initially, this shell thickening was attributed to the presence

of high levels of suspended sediment in the water (Key *et al.*, 1976). However, the incidence of abnormal shell growth in oysters also coincided with the increased use of organotin-based antifouling paints. Although such paints had been on the market since the 1960s, their use only became widespread in the 1970s with the development of self-polishing copolymer paints (Anderson & Dalley, 1986).

At the same time as British oyster growers were having problems with cultivation of *C. gigas*, the French were also concerned about the decline in their oyster fishery. It was thought that the presence of small boats coated with tributyltin (TBT)-based antifouling paints, close to oyster cultivation sites was responsible for the poor growth and shell thickening of *C. gigas* in France (Alzieu *et al.*, 1980). Consequently, in 1982, the French government banned the use of organotin compounds on boats under 25 m in length.

In the UK, field surveys and laboratory studies were carried out by the Ministry of Agriculture, Fisheries and Food (MAFF) during 1982–1985. These showed that TBT could be detected in the water at a number of locations around England and Wales, and that its presence coincided with shell thickening and reduced meat yield in *C. gigas* grown at these sites (Waldock & Miller, 1983; Thain & Waldock, 1986; Waldock, 1986). Furthermore, TBT produced the same effect under laboratory conditions (Waldock & Thain, 1983; Thain, 1986; Thain & Waldock, 1986).

As a result of this work, the UK Government took measures to reduce the environmental impact of TBT from antifouling paints (see Department of the Environment, 1986). In 1986, under the Control of Pollution Act (1974), the retail sale of organotin paints was restricted to those copolymer paints which contained less than 7.5% tin in the dry film, and those free-association paints which contained less than 2.5% tin. An environmental quality target (EQT) for TBT of 20 ng litre⁻¹ was set for estuarine and coastal waters (as, at that time, the level of 20 ng litre⁻¹ was thought likely to give adequate protection to marine species).

In 1986 MAFF and the Department of the Environment jointly initiated an extensive programme of monitoring concentrations of TBT in shellfish, sediment and water. The results of the 1986 field survey showed that the EQT of 20 ng litre⁻¹ was exceeded at more than half of the 40 locations sampled (Waldock *et al.*, 1987a), including the majority of the sites where oysters had historically been cultivated. High concentrations of TBT in water coincided with the presence of small boats. In the River Crouch, in Essex, oysters accumulated TBT to levels exceeding 1 µg g⁻¹ wet weight and growth was poor in terms of both meat weight and shell thickening (Waldock *et al.*, 1987b).

It was clear from the results of the 1986 monitoring programme that the UK Government's measures had been ineffective in reducing environmental

levels of TBT to meet the EQT of 20 ng litre^{-1} . Not only were TBT-painted small boats giving rise to unacceptably high levels of TBT in the water, but fish-farm cages coated with TBT-based antifoulant were also shown to be a major source of TBT in certain sea lochs in Scotland (Davies *et al.*, 1987). In addition, laboratory studies during 1985–1986 proved TBT to be even more toxic than at first thought. Thain *et al.* (1987) showed that shell thickening in *C. gigas* could be induced by TBT at levels of 20 ng litre^{-1} . Even lower concentrations of TBT (approximately 2 ng litre^{-1}) were shown to induce imposex (the presence of male characteristics in the female) in the dogwhelk *Nucella lapillus*, and so impair reproduction (Bryan *et al.*, 1986).

In 1987, as a result of the accumulated evidence from field and laboratory studies, the UK Government implemented controls, under the Food and Environment Protection Act (1985), to limit the sale and use of TBT-based antifouling paints. These were:

- (i) All antifoulants were to be treated as pesticides, and so could only be sold after approval by the Advisory Committee on Pesticides (an independent committee of experts).
- (ii) Products containing triorganotins were banned for use on vessels less than 25 m in length and for use on fish-farming equipment.
- (iii) Triorganotin paints could only be sold wholesale and in drums containing 20 litres or more.

In 1988, in response to the accumulated scientific evidence, including the findings of Bryan *et al.* (1986) and Thain *et al.* (1987), the Government set an environmental quality standard (EQS) for TBT in seawater of 2 ng litre^{-1} .

The 1987 legislation controlled both the sale of TBT-based paints and their use on boats less than 25 m in length, although they were not banned from use on large vessels. This was because field studies undertaken in 1986–1987 had shown that inputs of TBT to the environment from ships were considerably less than those from yachts (Waldock *et al.*, 1988). The input of TBT from ships is currently under review, and in particular the input associated with dry-docking practices is under investigation.

Since 1987, MAFF has continued with its programme of monitoring TBT in shellfish, sediment and water, in order to assess the effectiveness of the 1987 legislation. Attention has been focused on six estuaries which have traditionally supported oyster fisheries: the Blackwater and Crouch in Essex, the Dart, Kingsbridge and Teign in Devon, and the Beaulieu in Hampshire. All of these estuaries except the Teign are characterized by the presence of large numbers of small boats. The Beaulieu site represents a worst-case situation, as the shellfish are grown in cages suspended from a pontoon which is surrounded by moorings. The estuary most extensively studied is the Crouch: shellfish have been relaid at eight different locations

along its length, so as to provide information on TBT concentrations and shellfish growth compared with distance from boat moorings.

Monitoring of TBT water concentrations in marinas and harbours has also continued since 1987. A new marina was opened in 1988 at Hythe, on Southampton Water, after the ban on use of TBT based-paints for small boats. Analysis of water samples collected in Hythe marina should indicate any illegal use of TBT that is taking place, as TBT in the water at that site could only be derived by leaching from painted boat hulls, and not from desorption of TBT from the sediment.

MATERIALS AND METHODS

Shellfish sites

Since 1986, Pacific oyster (*Crassostrea gigas*) and mussel (*Mytilus edulis*) spat have been relaid each year at 13 estuarine sites. Eight of the sites (Holliwell Point, Holliwell Buoy, Roach Mouth, Bush Shore, Burnham, Creeksea, Bridgemarsh, Fambridge) are spaced at 3 km intervals along the River Crouch in Essex (Fig. 1). The other sites are: West Mersea on the River Blackwater, Essex; Blackness Point on the River Dart, Devon; Frogmore on the Kingsbridge estuary, Devon; Arch Brook on the River Teign, Devon; and Bucklers Hard on the Beaulieu River, Hampshire (Fig. 1).

Oyster spat of 1–2.5 g weight was supplied from a commercial hatchery in March each year. Mussels (1.0 g) were either obtained from a hatchery or were collected at Portwrinkle in Cornwall, an area which is not contaminated with TBT.

The method of relaying the shellfish followed the guidelines published by the MAFF shellfish cultivation unit (Spencer & Gouch, 1978; Spencer *et al.*, 1985). In March or April each year, approximately 200 oysters and 200 mussels were put into Netlon cages (6 mm mesh) and placed on trestles on the foreshore at the level of low water spring tides. Stocking density did not exceed 1 g cm^{-2} and the cages were cleaned of fouling organisms each month. Since samples of 20 oysters and 20 mussels were removed from the cages each month, the shellfish rarely outgrew the space available. However, where necessary the animals were moved on to larger cages with a broader mesh (15 mm). The exception to this strategy was the site at Bucklers Hard, on the Beaulieu River, where intertidal deployment was impossible. At this site animals were held in cages suspended from a pontoon, and so were kept fully submerged throughout the tidal cycle.

Samples of 20 oysters and 20 mussels were taken monthly from March/April to November/December from each of the shellfish sites. Water

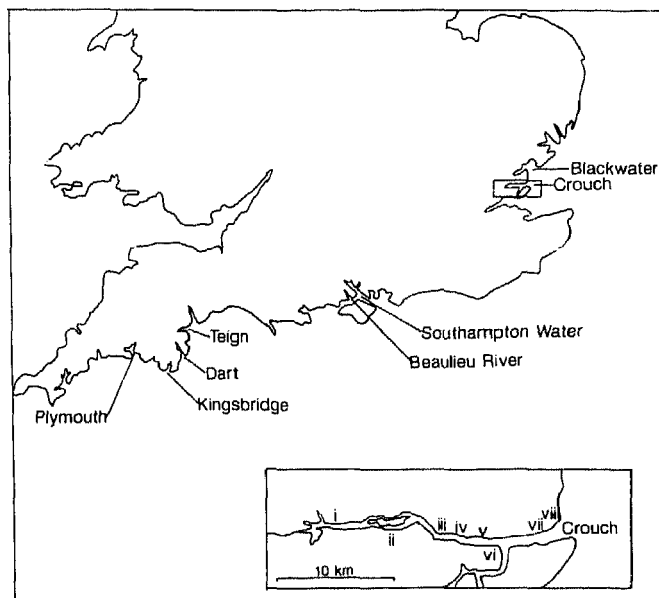


Fig. 1. Monitoring sites in southern England. i, Fambridge; ii, Bridgemarsh; iii, Creeksea; iv, Burnham; v, Bush Shore; vi, Roach Mouth; vii, Holliwell Buoy; viii, Holliwell Point.

samples (2 litres) were taken from 10 cm below the surface of the water, and surface skims of sediment were also collected monthly from the shellfish sites. For full details of sample collection and storage, see Waldock *et al.* (1989).

Growth performance of the oysters and mussels was monitored by measuring the increases in whole weight and meat weight (both on a wet weight basis). In the case of *Crassostrea gigas* the shell thickness index was also determined. The index is obtained by dividing the length of the upper shell valve by its thickness (Alzieu *et al.*, 1982; Waldock & Thain, 1983; Thain *et al.*, 1987).

Water, sediment, oyster and mussel tissues were analysed for organotin compounds using the methods of Waldock *et al.* (1989). Organotin compounds present in a sample were extracted with solvent, converted to their hydrides with sodium borohydride and then analysed by gas chromatography, using a flame photometric detector. The method is based on one developed by Matthias *et al.* (1986).

Harbours and marinas

Water samples were also taken from harbours and marinas and analysed for organotin compounds using the above method. Dartmouth Marina on the

River Dart, Teignmouth Harbour on the River Teign and Sutton Marina near Plymouth, Devon (Fig. 1) have all been sampled on a monthly basis (April–November) since 1986. Water samples have also been collected monthly from Hythe Marina on Southampton Water (Fig. 1) since it opened in 1988.

RESULTS

Concentrations of tributyltin in water samples

Shellfish sites

Mean concentrations of TBT in water have decreased at all of the shellfish sites monitored since 1986 (Table 1). In general the water concentrations observed in 1989 are one-third to one-quarter of those recorded in 1986. Bucklers Hard was found to be the most contaminated of the shellfish sites during the period 1986–1989.

TABLE 1
Mean Summer (May–September) Concentrations of Tributyltin in the Water of UK Estuaries, Marinas and Harbours, 1986–1989 (ng litre⁻¹)

Estuary/Water body	Location	1986	1987	1988	1989
	Shellfish sites				
Crouch	Fambridge	15±8	33±27	21±8	13
	Bridgemarsh	22±12	17±12	13	8
	Creeksea	35±17	17±9	22±14	8±2
	Burnham	45±17	31±18	23±18	11±4
	Bush Shore	26±9	22±15	13±5	8
	Roach Mouth	26±12	18±13	15±12	8±2
	Holliwell Buoy	11	26±23	10±4	3±2
Blackwater	Holliwell Point	16	6±5	6±5	2
	West Mersea	38±21	36±29	76±43	25
Dart	Blackness Point	38±33	13±4	13±5	8±3
Kingsbridge	Frogmore	15±6	11±6	51±95 ^b	5±2
Teign	Arch Brook	12±10	7±6	6±2	6±4
	Marinas and harbours				
Plymouth	Sutton Marina	1160±84	882±323	274±79	266±134
Dart	Dart Marina	95±66	85±32	21±4	16±6
Kingsbridge	Salcombe	117±84	62±71	30±18	21±15
Teign	Teignmouth	22±24	23±25	19±12	25±15
Beaulieu River	Bucklers Hard*	93±45	1090±1850 ^a	82±9	25±7
Southampton Water	Hythe Marina			1960±2470 ^c	93±62

* Also a shellfish site.

^a 263±130 without June value. ^b 8±2 without August value. ^c Mean = 728 without May value. SD only given where $n \geq 4$.

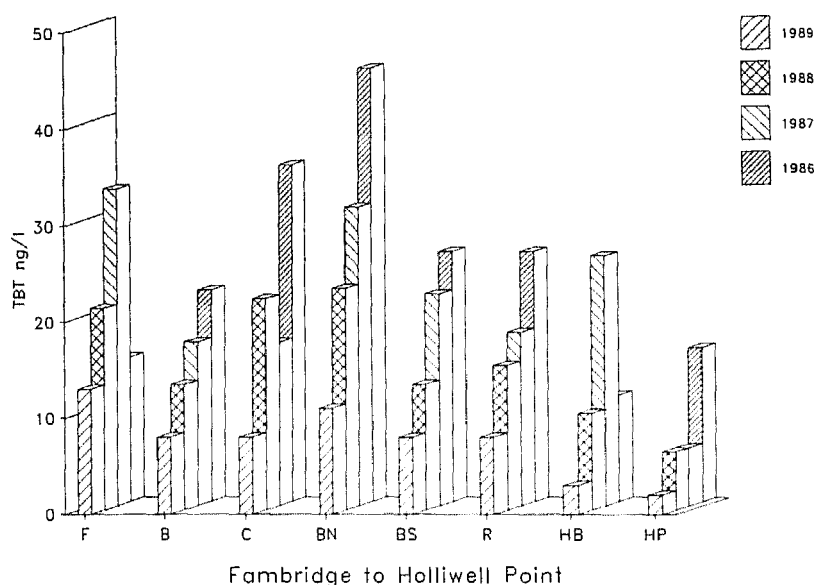


Fig. 2. Mean (May–September) concentrations of TBT in water at eight sites on the River Crouch (1986–1989). F, Fambridge; B, Bridgemarsh; C, Creeksea; BN, Burnham; BS, Bush Shore; R, Roach Mouth; HB, Holliwel Buoy; HP, Holliwel Point (mouth of the estuary).

Figure 2 shows annual mean TBT water concentrations from 1986 to 1989 for the eight sites along the River Crouch. Concentrations were lowest at the mouth of the estuary at Holliwel Point, and highest in the middle of the estuary at Burnham, where there is a high density of boat moorings. In 1986 the level of TBT at Holliwel Point was 16 ng litre^{-1} , while at Burnham it was 45 ng litre^{-1} . Water concentrations have also been high in the upper estuary at Fambridge, which is close to a small marina. From 1987 to 1989 the concentrations at Fambridge were virtually the same as at Burnham ($33\text{--}13 \text{ ng litre}^{-1}$ and $31\text{--}11 \text{ ng litre}^{-1}$, respectively).

Since 1986 there has been a decrease in mean water concentrations at each of the sites on the Crouch, and by 1989 TBT levels were less than 10 ng litre^{-1} at all but two of the sites. At Burnham the 1989 concentration (11 ng litre^{-1}) was one-quarter of that recorded in 1986, and at Holliwel Point in 1989 the level of TBT was only 2 ng litre^{-1} (i.e. the same as the EQS).

Harbours and marinas

Concentrations of TBT in water in Sutton Marina, Dart Marina, Salcombe Harbour and Bucklers Hard by 1989 were approximately a quarter of those recorded in 1986 (Table 1). Sutton Marina still remains heavily contaminated with TBT, however, as in 1988 and 1989 the mean summer TBT concentration in water was approximately $270 \text{ ng litre}^{-1}$. At the other

three sites the concentrations of TBT in water were down to around 20 ng litre⁻¹ in 1989.

The levels of TBT in Teignmouth Harbour have remained steady, 19–25 ng litre⁻¹ over the entire period 1986–1989. Teignmouth is different to the other marina and harbour sites in that, although busy, it is used mainly by ships rather than small boats. This illustrates the lesser impact of ships >25 m in length compared with that of smaller vessels.

Extremely high concentrations of TBT have been found in Hythe Marina, e.g. in May 1988 over 5.6 µg litre⁻¹ was recorded. Since this was a new marina in 1988, the results indicate illegal use of TBT antifouling paint in that year. The average summer concentration observed in 1989 was considerably lower than that seen during 1988.

Concentrations of tributyltin in *Crassostrea gigas*

The values in Table 2 are means of June, July and August TBT concentrations in *C. gigas* tissue. Typically, tissue concentrations with a mean of 1.0 µg g⁻¹ would have a range of 0.9–1.1 µg g⁻¹. In 1986 the results for oyster tissues were obtained using a slightly different method to that used in subsequent years. A comparison of the two methods (Waldock *et al.*, 1989) showed that the earlier method yielded TBT values which were approximately 82% of the values obtained by using the later method. Consequently, the concentrations of TBT in oysters shown in Table 2 for 1986 have been corrected to accord with later values.

TABLE 2
Mean Summer (June–August) Concentrations of Tributyltin in *Crassostrea gigas*, 1986–1989
(µg g⁻¹ wet weight)

Estuary	Site	1986	1987	1988	1989
Crouch	Fambridge	1.61	1.64	0.62	0.36
	Bridgemarsh	1.20	1.46	0.44	0.33
	Creeksea	1.49	1.73	0.61	0.38
	Burnham	1.24	1.57	0.50	0.45
	Bush Shore	0.74	1.26	0.34	0.31
	Roach Mouth	0.80	0.98	0.24	0.27
	Holliwell Buoy	0.37	0.56	0.17	0.11
	Holliwell Point	0.18	0.28	0.08	0.08
Blackwater	West Mersea	2.26	2.18	1.34	0.65
Dart	Blackness Point	0.88	1.35	0.50	0.26
Kingsbridge	Frogmore	1.39	1.44	0.48	0.21
Teign	Arch Brook	0.30	0.49	0.25	0.13
Beaulieu	Bucklers Hard	6.35	3.65	5.60	1.28

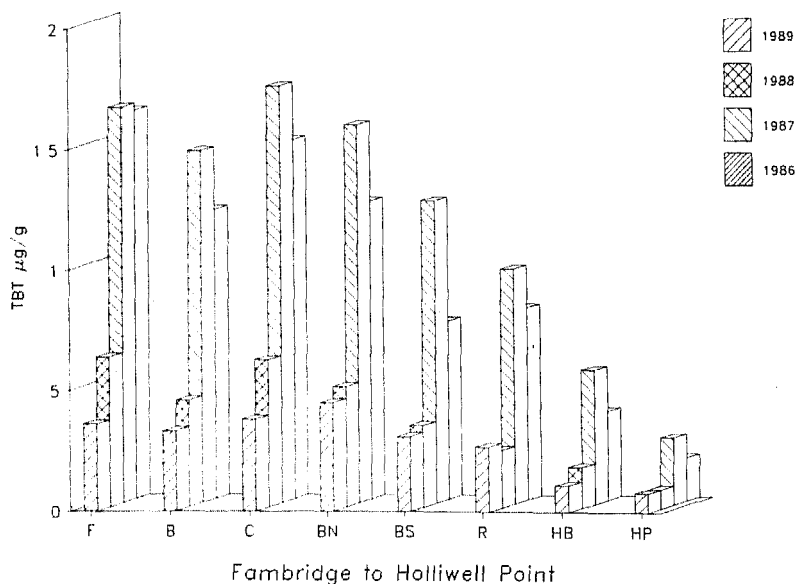


Fig. 3. Mean (June–August) concentrations of TBT in *Crassostrea gigas* at eight sites on the River Crouch (1986–1989). F, Fambridge; B, Bridgemarsh; C, Creeksea; BN, Burnham; BS, Bush Shore; R, Roach Mouth; HB, Holliwell Buoy; HP, Holliwell Point (mouth of the estuary).

Tributyltin concentrations in *C. gigas* have decreased at all sites since 1986 (Table 2). The 1989 TBT levels are approximately one-third to one-quarter of those measured in 1986. However, at Bucklers Hard in 1989 oysters still contained concentrations of TBT over $1 \mu\text{g g}^{-1}$ wet weight.

Figure 3 shows concentrations of TBT in *C. gigas* relaid along the River Crouch. The concentration of TBT in oysters follows a similar pattern to that for water, e.g. in 1989 the concentration in oysters at the mouth of the estuary was low ($0.08 \mu\text{g g}^{-1}$ wet weight), whilst in the middle of the estuary at Burnham, and close to the marina at Fambridge, the tissue concentrations were much higher, ($0.45 \mu\text{g g}^{-1}$ wet weight and $0.36 \mu\text{g g}^{-1}$ wet weight, respectively). The most marked decrease in tissue concentrations was seen between 1987 and 1988 (Fig. 3), following the 1987 ban on use of TBT-based paint on small boats.

The concentrations of TBT found in oyster tissues are generally up to 50 000 times greater than in the water to which the oysters are exposed.

Concentrations of tributyltin in *Mytilus edulis*

Concentrations of TBT in *Mytilus edulis* have also decreased at all sites, so that the 1989 values are one-third to one-quarter of those recorded in 1986

TABLE 3
Summer Concentrations of Tributyltin in *Mytilus edulis*, 1986–1989 ($\mu\text{g g}^{-1}$ wet weight)

Estuary	Site	1986	1987	1988	1989
Crouch	Fambridge	0.64	0.75	0.34 ^b	0.16
	Bridgemarsh	0.66	0.44	0.30 ^b	0.15
	Creeksea	1.01	0.61	0.46 ^b	0.29
	Burnham	0.98	1.04 ^b	0.38 ^d	0.29
	Bush Shore	0.75	0.47	0.27 ^b	0.17 ^a
	Roach Mouth	0.45	0.41	0.19 ^b	0.12
	Holliwell Buoy	0.30	0.35	NS	0.07
	Holliwell Point	0.17	0.11	0.11 ^b	0.05
Blackwater	West Mersea	1.06	0.82	0.32 ^c	0.35 ^b
Dart	Blackness Point	0.60	0.41	0.20	0.11
Kingsbridge	Frogmore	0.93	0.58	0.19	0.10
Teign	Arch Brook	0.33	0.23	0.09	0.07
Beaulieu	Bucklers Hard	2.57	3.22	NS	0.81

All values are for August samples except: ^a June; ^b July; ^c September; ^d November.
NS = not sampled.

(Table 3). In general, mussels accumulate less TBT than oysters, and mussel tissue concentrations were approximately half those for oysters at the same site. Bioaccumulation factors for TBT in these mussels are generally about 20 000.

Figure 4 shows TBT concentrations for mussels re-laid along the River Crouch. The pattern of TBT accumulation in mussels is similar to that for oysters, i.e. concentrations are lowest at the mouth of the estuary at Holliwell Point (e.g. $0.05 \mu\text{g g}^{-1}$ wet weight in 1989) and highest in the middle of the estuary near Burnham (e.g. $0.29 \mu\text{g g}^{-1}$ wet weight in 1989). The most noticeable decrease in concentrations of TBT in *M. edulis* occurred between 1987 and 1988, as was the case with oysters.

Concentrations of tributyltin in sediments

During 1986, samples of sediment were collected weekly from sites in the Crouch. Analysis of the samples showed considerable variation in the concentrations of TBT from week to week. In certain estuaries such as the River Crouch, the sediment is very mobile. Sampling sites may be subject to scouring, so that contaminated sediment is removed, or to deposition of sediment which may be more or less contaminated than the surface layer. Consequently, sediments can only be used to give a general indication of TBT contamination within an estuary where the sediment is very mobile.

Concentrations of TBT in sediment collected in the summer from the

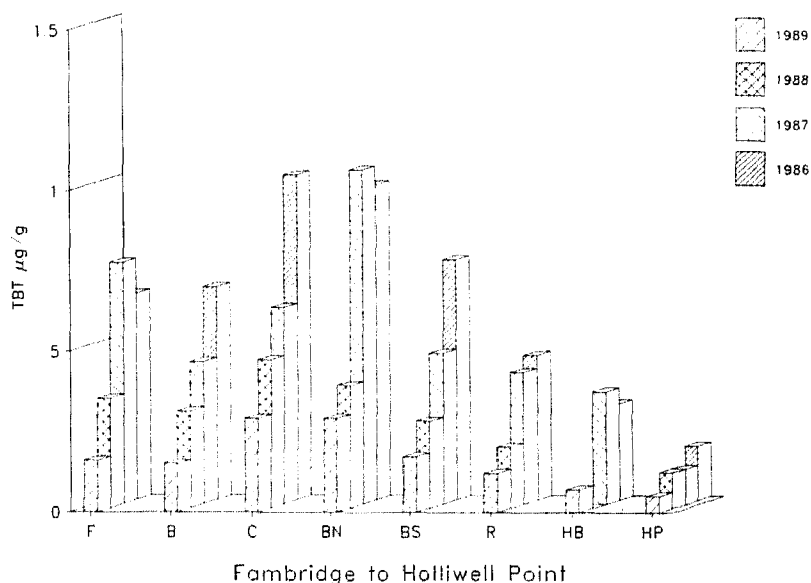


Fig. 4. Summer concentrations of TBT in *Mytilus edulis* at eight sites on the River Crouch (1986–1989). F, Fambridge; B, Bridgemarsh; C, Creeksea; BN, Burnham; BS, Bush Shore; R, Roach Mouth; HB, Holliwel Buoy; HP, Holliwel Point (mouth of the estuary).

different shellfish sites, during the period 1986–1989, are given in Table 4. No clear trend in the data from year to year can be seen, except for samples from Bucklers Hard where TBT concentrations have decreased since 1987, when they were exceptionally high at $10.8 \mu\text{g g}^{-1}$ dry weight.

The shellfish sites may be divided into four categories according to the level of TBT contamination of the sediment:

- (i) Lightly contaminated sites (Holliwel Point, Holliwel Buoy, Roach Mouth, Arch Brook), which are remote from boat moorings and have TBT concentrations of $<0.01\text{--}0.05 \mu\text{g g}^{-1}$ dry weight.
- (ii) Medium contaminated sites (Fambridge, Bridgemarsh, Creeksea, Blackness Point, Frogmore), which are closer to moorings and have TBT concentrations in the range $0.06\text{--}0.2 \mu\text{g g}^{-1}$ dry weight.
- (iii) Highly contaminated sites (Burnham, Bush Shore, West Mersea) within high density mooring areas, with TBT concentrations in the range $0.3\text{--}1 \mu\text{g g}^{-1}$ dry weight.
- (iv) Sites where the sediment contains paint particles and consequently has TBT concentrations of $>1 \mu\text{g g}^{-1}$ dry weight (Bucklers Hard).

Figure 5 shows TBT sediment concentrations at the eight shellfish sites in the Crouch from 1986 to 1989. The highest concentrations occurred at Burnham ($0.36 \mu\text{g g}^{-1}$ dry weight in 1986) and the lowest at the mouth of the

TABLE 4
Summer Concentrations of Tributyltin in Sediments, 1986–1989 ($\mu\text{g g}^{-1}$ dry weight)

Estuary	Site	1986	1987	1988	1989
Crouch	Fambridge	0.08	0.03	0.02	0.10
	Bridgemarsh	0.15	0.04	0.08	0.05
	Creeksea	0.04	0.11 ^a	0.07	0.04
	Burnham	0.36	0.15 ^b	0.31	0.10
	Bush Shore	0.07	0.05	0.27	0.07
	Roach Mouth	0.05	0.02	0.02	0.04
	Holliwell Buoy	<0.01	0.01	0.02	0.02
	Holliwell Point	0.01	0.01	0.05	<0.01
	Blackwater	West Mersea	0.66	0.26	0.15
Dart	Blackness Point	0.08	0.19	0.17	0.07
Kingsbridge	Frogmore	0.06 ^b	0.03	0.07	0.04
Teign	Arch Brook	0.05 ^b	0.02	0.03	0.02
Beaulieu	Bucklers Hard	4.6	10.8	1.1	0.23

All values are for August samples except: ^a July; ^b September.

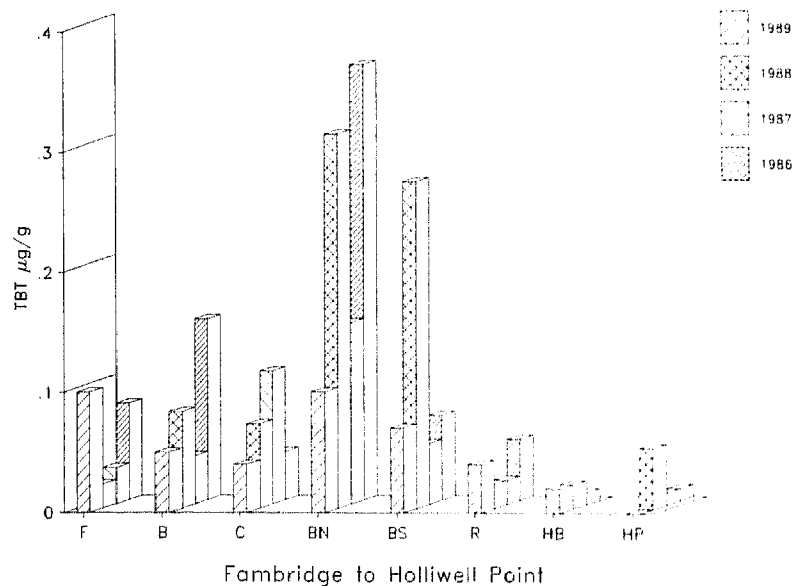


Fig. 5. Summer concentrations of TBT in sediment at eight sites on the River Crouch (1986–1989). F, Fambridge; B, Bridgemarsh; C, Creeksea; BN, Burnham; BS, Bush Shore; R, Roach Mouth; HB, Hollwell Buoy; HP, Hollwell Point (mouth of the estuary).

estuary at Holliwell Point ($0.01 \mu\text{g g}^{-1}$ dry weight). However, by 1989 the movement of contaminated and/or uncontaminated sediments appeared to have caused a more even distribution of TBT in the estuary, and the marked elevations previously seen at Burnham were not apparent.

The concentrations of TBT in the surface sediments in the Crouch were approximately 1000–6000 times greater than those measured in the water.

Shell thickness index in *Crassostrea gigas*

Shell thickness indices for *C. gigas* have increased at all sites since 1986, except at Bucklers Hard (Table 5). The most marked improvement in shell shape (i.e. growth of thin shells) was between 1987 and 1988. Normal shaped oysters (those with thin shells, shell thickness index > 10) can now be grown in the three Devon estuaries, the Blackwater, and at Holliwell Buoy and Holliwell Point near the mouth of the River Crouch. At Bucklers Hard, the site most contaminated with TBT, oysters in 1989 had a shell thickness index of < 7 .

In the upper and middle regions of the River Crouch in 1989, shell thickness indices were close to 10 (Fig. 6) and corresponded to TBT tissue concentrations of $0.3 \mu\text{g g}^{-1}$ wet weight to $0.5 \mu\text{g g}^{-1}$ wet weight and water concentrations of approximately 10 ng litre^{-1} . *Crassostrea gigas* grown near the mouth of the estuary, at Holliwell Buoy and Holliwell Point, were much thinner-shelled and had shell thickness indices close to 20, which

TABLE 5
August Shell Thickness Index Values for *Crassostrea gigas* 1986–1989

Estuary	Site	1986	1987	1988	1989
Crouch	Fambridge	4.39	4.85	6.85	9.82
	Bridgemarsh	4.78	4.67	8.10	10.2
	Creeksca	5.14	5.29	8.07	9.83
	Burnham	4.95	5.04	7.90	10.2
	Bush Shore	5.06	5.24	9.83	NS
	Roach Mouth	5.39	5.29	9.00	8.62
	Holliwell Buoy	9.63	6.48	NS	19.8
	Holliwell Point	12.4	9.64	23.3	21.0
Blackwater	West Mersea	4.95	3.96	6.87	14.9
Dart	Blackness Point	10.6	5.98	12.5	14.7
Kingsbridge	Frogmore	7.56	5.41	9.26	13.1
Teign	Arch Brook	10.6	12.4	25.7	18.9
Beaulieu	Bucklers Hard	3.21	8.06	4.34	6.73

NS = not sampled.

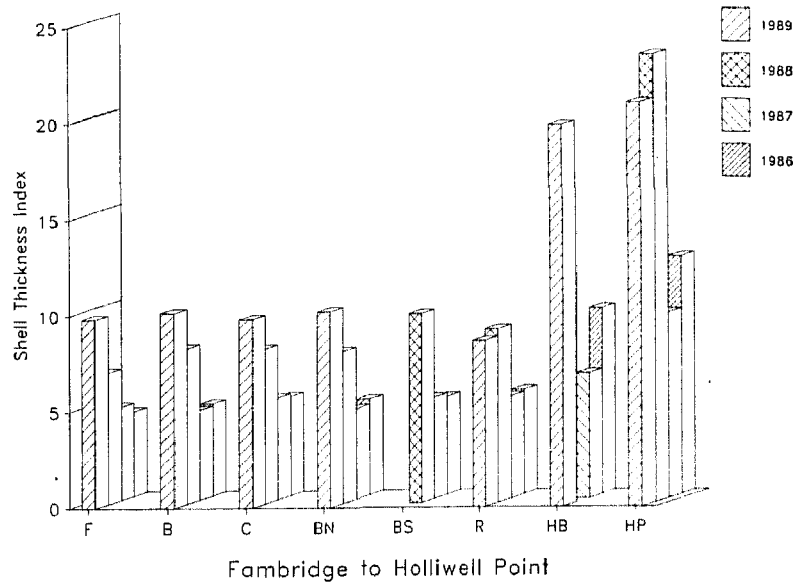


Fig. 6. August shell thickness index of *Crassostrea gigas* at eight sites on the River Crouch (1986–1989). F, Fambridge; B, Bridgemarsh; C, Creeksea; BN, Burnham; BS, Bush Shore; R, Roach Mouth; HB, Holliwell Buoy; HP, Holliwell Point (mouth of the estuary).

TABLE 6
August Meat Weight Values (g wet wt) for *Crassostrea gigas*, 1986–1989

Estuary	Site	1986	1987	1988	1989
Crouch	Fambridge	1.29	0.70	2.59	5.66
	Bridgemarsh	1.26	0.67	2.62	2.84
	Creeksea	0.97	0.84	2.23	2.90
	Burnham	0.77	0.49	2.39	2.77
	Bush Shore	1.24	1.24	2.44	NS
	Roach Mouth	1.18	1.14	2.50	5.92
	Holliwell Buoy	1.39	1.44	NS	5.96
	Holliwell Point	2.72	2.06	4.11	9.28
Blackwater	West Mersea	1.00	0.97	2.53	3.65
Dart	Blackness Point	1.56	0.89	3.19	7.09
Kingsbridge	Frogmore	1.77	1.32	4.53	8.07
Teign	Arch Brook	1.56	1.24	5.22	6.23
Beaulieu	Bucklers Hard	0.37	0.12	0.95	1.24

NS = not sampled.

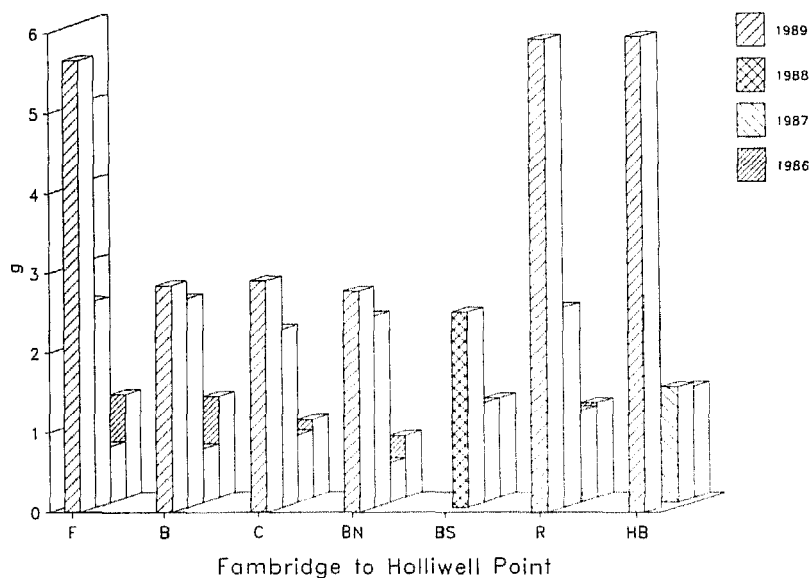


Fig. 7. August meat weight values (g wet wt) of *Crassostrea gigas* at eight sites on the River Crouch (1986–1989). F, Fambridge; B, Bridgemarsh; C, Creeksea; BN, Burnham; BS, Bush Shore; R, Roach Mouth; HB, Holliwell Buoy; HP, Holliwell Point (mouth of the estuary).

corresponded to TBT body burdens of $0.1 \mu\text{g g}^{-1}$ wet weight, and water concentrations of 2 to 3 ng litre^{-1} .

Meat weight of *Crassostrea gigas*

Oyster production, in terms of meat weight, has improved at every site since 1986, although in 1989 oysters relaid at Bucklers Hard still had a very poor growth rate (Table 6). At all of the sites the most noticeable improvement in meat weight was between 1987 and 1988, as was the case for shell thickness index. Nevertheless in 1989, on the River Crouch, meat yield in oysters from the middle of the estuary was still reduced compared to the upper estuary and the mouth (Fig. 7).

DISCUSSION

Contamination of the marine environment with tributyltin (TBT) has been shown to occur in many countries. The levels of TBT found in large marinas in Britain during the 1980s sometimes exceeded $1 \mu\text{g litre}^{-1}$, but were generally of the order of hundreds of nanograms per litre (this study; Cleary & Stebbing, 1987; Waldock *et al.*, 1987a) and were similar to those reported for marinas in France (Alzieu *et al.*, 1989; 1990), the Netherlands (Laane *et*

al., 1990), Spain (Tolosa *et al.*, 1990), Greece (Fytianos & Samanidou, 1990), Turkey (Kubilay *et al.*, 1990), the United States (see Hall, 1988), Australia (Batley *et al.*, 1989a) and New Zealand (King *et al.*, 1989).

High levels of TBT have also been shown to occur in Leghorn Harbour, Italy; however, in this case TBT was derived from both power station effluent and dry-docking activities, as well as from small boats (Bacci & Gaggi, 1989).

Harbour and estuarine sediments are contaminated with TBT in many parts of the world. In Britain, TBT concentrations vary from $0.06 \mu\text{g g}^{-1}$ dry weight to $> 1 \mu\text{g g}^{-1}$ dry weight in sediments from marinas and areas with high densities of boat moorings, or in sediments close to boatyards (this study; Ashby & Craig, 1989). Similar sediment concentrations have been found in marinas in the United States (Makkar *et al.*, 1989; Matthias *et al.*, 1988; Rice *et al.*, 1987) and New Zealand (King *et al.*, 1989). In the Sado estuarine system, Portugal, contamination of sediments is a result of discharges of TBT from shipyards in the area (Quevauviller *et al.*, 1989), and in Egypt the levels of TBT in sediments along the Alexandria coast are derived primarily from TBT-based paints used on ships (Aboul Dahab, 1990).

The effects of tributyltin on the cultivation of the Pacific oyster *Crassostrea gigas* in England and France have been well documented (this study; Alzieu *et al.*, 1980; Alzieu & Portmann, 1984; Thain & Waldock, 1986; Waldock, 1986). *Crassostrea gigas*, grown in estuaries contaminated with TBT, accumulate TBT and suffer from reduced meat yields and shell thickening. In Australia, Batley *et al.* (1989b) reported that, when Sydney oysters (*Saccostrea commercialis*) were grown close to high densities of boats, they had a TBT tissue concentration of $0.13 \mu\text{g Sn g}^{-1}$ ($0.33 \mu\text{g TBT g}^{-1}$), their shells were deformed and their meat weights reduced. Accumulation of TBT has also been shown to occur in *Crassostrea virginica* cultivated close to a marina in Sarah Creek, Chesapeake Bay (Rice *et al.*, 1987). In an extensive study of the United States, Wade *et al.* (1988) reported the presence of TBT in oysters (*Crassostrea virginica* and *Ostrea sandwichensis*) grown on the Atlantic, Gulf of Mexico and Pacific coasts.

Mussels, like oysters, also accumulate TBT, although *Mytilus edulis* exhibits a lower bioconcentration factor than *C. gigas* (this study). As mussels are fairly widespread, are sedentary, and are good accumulators of TBT, they are useful as bioindicators of TBT contamination. Accumulation of TBT in *M. edulis* has been found to occur in samples from Alaska (Short & Sharp, 1989), California (Salazar & Salazar, 1988; Wade *et al.*, 1988), the north-east coast of the United States (Page *et al.*, 1990a; Wade *et al.*, 1988), Denmark (Zuolian & Jensen, 1989) and Norway (Page & Widdows, 1991). In addition Page *et al.* (1990b) have shown that TBT is concentrated in the

tissues of *Mytilus galloprovincialis* sampled along the Mediterranean coast of France and Italy. Mussels (*Arca zebra*) from Hamilton harbour, Bermuda were also found to accumulate TBT (Page & Widdows, 1991).

It is thus clear that contamination of the environment with TBT occurs in many parts of the world. Reports of TBT contamination of water, sediment or shellfish have come from Europe, North America, Australia, New Zealand and North Africa. The Japanese have also published data on the accumulation of TBT in yellowtails reared in marine farms (Sasaki *et al.*, 1988). The degree of contamination in the coastal waters of Asia, South America and much of Africa is as yet unknown.

In many of the developed countries legislation has now been enacted to control the sale and/or use of organotin-based antifoulants. France was the first country to institute control over the use of TBT-based paints; in 1982 the French government banned the use of organotin paints on boats of less than 25 m in length (see Alzieu, 1991). In Italy, since 1985, it has been illegal to use organotin biocides in industrial cooling waters which are discharged to shellfish farming areas (see UNEP, 1989). Switzerland and Germany had both prohibited the use of TBT-based antifouling paints in freshwater by 1987 (Champ & Pugh, 1987). In Britain in 1987, TBT antifoulants were banned from use on boats under 25 m in length, and on fish-farming equipment (Abel *et al.*, 1987). Eire, Norway, Sweden, the Netherlands, Canada, Australia (New South Wales and Tasmania) and New Zealand have also banned the use of TBT-based paints on vessels under 25 m in length (Dahl & Blanck, 1990; Dalley, 1989; Laane *et al.*, 1990). In several of these countries small vessels constructed from aluminium are exempt from the ban (see Dalley, 1989). In the United States, in 1988, TBT was prohibited for use on all boats less than 25 m in length that have non-aluminium hulls (Anon, 1989), and only Environmental Protection Agency-certified paints may be used on other vessels (Schatzberg, 1990). In Japan there is little statutory control of TBT-based antifoulants as yachting is not widespread, although the fish-farming associations have agreed that only copolymer paints with a low release rate of TBT should be used on fish cages (Dalley, 1989).

The present study shows that controls instituted in Britain in 1987 on the use of TBT-based paints for small boats and mariculture equipment have been very effective in reducing water concentrations of TBT, with a concomitant improvement in oyster growth. A *C. gigas* fishery on the River Blackwater, on the east coast of England, which had been closed since the 1970s, was reopened in late 1987 and was producing oysters of near-marketable size by the end of 1988. Cleary (1991) also reports significant reductions in TBT water concentrations since 1987 at coastal and estuarine sites in the south-west of England.

Legislation in other countries has been effective in reducing TBT contamination of the environment. Since 1982 in the oyster growing areas of France, the concentrations of TBT in water have decreased and are now generally $< 5 \text{ ng litre}^{-1}$ (Michel & Alzieu, 1990). TBT contamination of oysters, *C. gigas*, has become much less and spatfall has greatly improved (Alzieu, 1991). In the USA, Valkirs *et al.* (1991) have found reductions in the levels of TBT in yacht harbours in San Diego Bay, which they attribute to the restriction of use of TBT-based paints on small boats.

There does appear to be a continuing and illegal use of TBT-based antifoulants in several countries. In Britain, water samples taken from Hythe Marina in 1988 had a mean TBT concentration of approximately $2 \mu\text{g litre}^{-1}$. As this marina was only opened in 1988, after the 1987 ban on use of TBT-based paints on small boats, the levels of TBT in the water cannot be accounted for by desorption of TBT from the sediment. Neither can the high levels of TBT be a result of the hosing down of boats coated with old layers of TBT-paint, as there are no boatyard facilities at Hythe Marina. The extremely high concentrations of TBT in the water seem most likely to have been the result of the presence of boats which had been recently (and illegally) painted with TBT-based antifoulants in 1988. In France, several years after the French ban on the use of TBT-based paints for small boats, Alzieu *et al.* (1989) reported that abnormally high inputs of TBT to the environment were occurring. This was particularly noticeable in Boyardville Marina on the Atlantic coast, where TBT concentrations of up to $1.5 \mu\text{g litre}^{-1}$ were recorded.

Although the legislation in Britain, which banned the use of TBT-based paints on small boats, has caused concentrations of TBT in estuaries and marinas to drop substantially since 1987, the environmental quality standard (EQS) of 2 ng litre^{-1} has only been achieved at one of the twelve open, estuarine sites (Hollwell Point on the River Crouch), and at none of the marina sites monitored in the present study. The levels of TBT at all of the monitoring sites in 1989 were sufficiently high to induce imposex in the dogwhelk *Nucella lapillus* (Bryan *et al.*, 1986), the most TBT-sensitive organism as yet reported.

In 1987 TBT-based antifoulants were also banned from use on mariculture equipment in Britain. Bailey & Davies (1991) observed that when dogwhelks were transplanted close to sites of mariculture in Loch Laxford, in Scotland, there was a deterioration in the dogwhelks' reproductive condition, indicating that TBT was still present in the water.

It is unlikely that the illegal use of TBT-based antifoulants can entirely account for the persistence of TBT in the water at all of the sites monitored in the present study. Also, as the majority of the sites are not associated with shipping activity, the source of the TBT cannot be ships, which are exempt

from the 1987 ban. Despite the fact that TBT has a relatively short half-life in water (approximately 6 days under optimum conditions (Waite *et al.*, 1989)), it has a half-life of almost 2 years in anaerobic sediments (Waldock *et al.*, 1990). It is probable that the TBT water concentrations at many of the sites are due to desorption of TBT from the sediments. Waldock *et al.* (1990) believe that contaminated sediments may prove to be a source of TBT for several years to come.

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