



Biological Indicators Used to Map Organotin Contamination from a Fishing Port, Killybegs, Ireland

DAN MINCHIN*§, BARBARA BAUER†, JÖRG OEHLMANN‡, ULRIKE SCHULTE-OEHLMANN‡ and COLM B. DUGGAN*

*Fisheries Research Centre, Department of the Marine, Abbotstown, Dublin 15, Ireland

†Institut für Spezielle Zoologie and Vergleichende Embryologie, Westfälische Wilhelms-Universität, Hüfferstraße 1, D-48149 Münster, Germany

‡Internationales Hochschulinstitut Zittau, Lehrstuhl für Umweltverfahrenstechnik, Markt 23, D-02763 Zittau, Germany

Contamination by organotins in areas of aquaculture and small boat activity about Ireland has generally decreased since legislation was introduced in 1987, but in many port areas contamination increased. The fishing port of Killybegs lies within a small sheltered inlet in Donegal Bay on the N-W Irish coast and is the main source of contamination for several kilometers. This area has been studied using imposex of dogwhelks, *Nucella lapillus*, since 1987. Dogwhelks are particularly sensitive to TBT and have become extinct within the Harbour, and remaining populations near the Harbour entrance are likely to die-out. Periwinkles, *Littorina littorea*, are less affected by TBT and have a condition, intersex, which was used to determine the relative levels of contamination within the Harbour in areas where dogwhelks no longer exist. Both indicator species compliment each other; but periwinkles, useful for areas where there is high contamination, can provide some unexpected results in areas of low contamination. For such areas dogwhelks are better indicators. Imposex and intersex values decline with distance from source; low levels of contamination were found 12 km from Killybegs port. Decrease of these biological indices with distance from source was generally consistent with TBT body burdens. Fishing vessels that are greater than 25 m are permitted to use organotin antifoulant paints and high levels of contamination are expected to remain in the immediate area of the Harbour for some years even if its use is discontinued. © 1997 Elsevier Science Ltd

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Following Irish restrictions on the usage of all organotins in paint formulations, including tributyltin

(TBT) on vessels less than 25 m, in 1987 (Anon., 1987), there was a general decrease in contamination in areas where TBT was formerly used on recreational vessels and on salmon-farm cage netting (Minchin *et al.*, 1987, 1995; Minchin, 1995). There is a world-wide trend of a decrease in TBT contamination in many areas (Bailey & Davies, 1991; Waite *et al.*, 1991; Dyrinda, 1992; Dowson *et al.*, 1993; Bryan *et al.*, 1993; Evans *et al.*, 1994, 1995; Tester & Ellis, 1995). However in port areas, TBT contamination remains high (Davies & Bailey, 1991; Uhler *et al.*, 1993; Minchin *et al.*, 1995, 1996). Under present Irish legislation larger fishing boats are permitted to use this substance. The nearby boatyards, which repaint these vessels, must also contribute to the contamination. TBT is toxic to a wide spectrum of organisms, and low concentrations can result in reproductive impairment in many marine snails (Deutsch & Brick, 1993; Wilson *et al.*, 1993). The dogwhelk, *Nucella lapillus*, a muricid snail, has been used as a standard biological indicator for the presence of TBT in several previous studies. There is evidence that at least 50% of the TBT concentrated in these snails is obtained through feeding (Bryan *et al.*, 1989). TBT may be present in the environment at low levels and can bioaccumulate in the organisms on which these predatory snails feed (Oehlmann, 1994; Stroben, 1994).

Killybegs Harbour is a long, narrow, sheltered and fully tidal inlet on the north-west coast of Ireland (Fig. 1). It is the main fishing port on this coast. Boatyards which service the fishing fleet, and piers at which the vessels berth, lie within half a kilometre on the western side of the Harbour. The numbers of vessels in port depend on weather and seasonal fishing activities; vessels may remain in port several days. A small number of coastal ships also visit this port. By the mid-1970s TBT antifouling was widely used by shipping in Ireland and it is likely that it has been used in Killybegs since about this time. The first direct evidence of TBT contamination in Killybegs Harbour was in a study in 1985 using the Pacific oyster, *Crassostrea gigas*,

§Author to whom correspondence should be addressed.

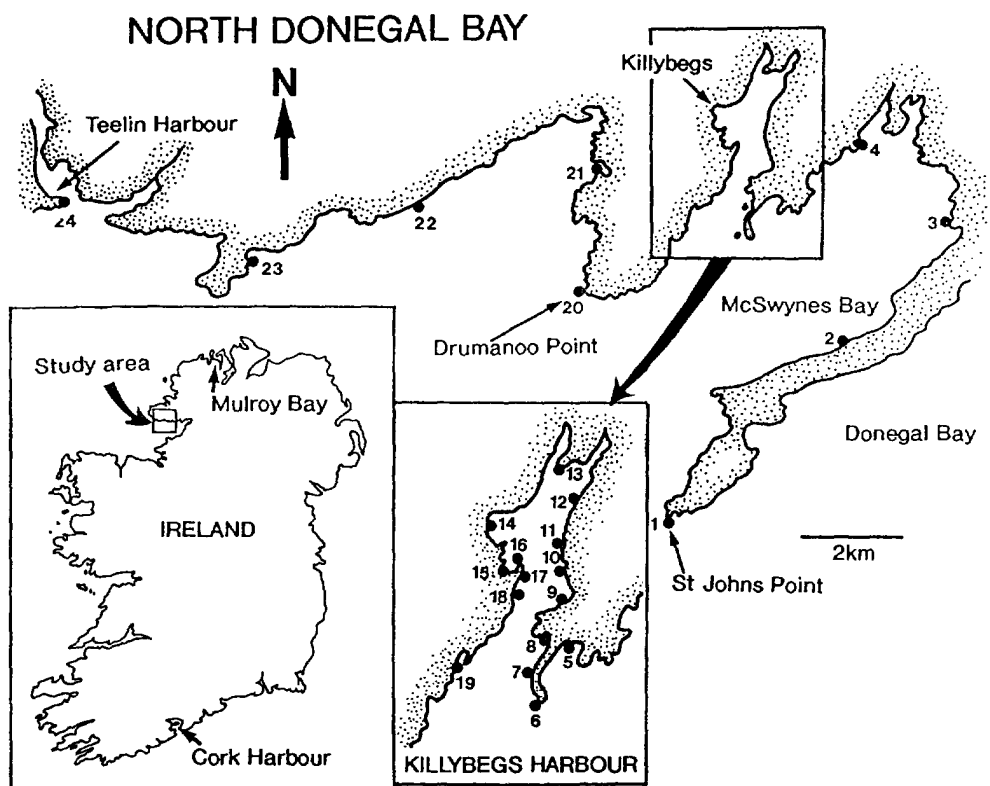


Fig. 1 Distribution of sampling stations where snails were collected. Sediment samples were taken half way between stations 11 and 16.

as an indicator. These developed thick distorted shells which contained a gel, as attributed to TBT contamination by Alzieu *et al.* (1982). The shell thickness index for these oysters after six months to September was 4.7, indicating an already high level of contamination at that time.

In 1987 dogwhelk imposex was used to determine relative levels of contamination about the Irish coast so as to provide a baseline for measuring changes (Duggan *et al.*, 1988). Stations established in this survey in the Killybegs region were again sampled in 1993 and these showed increases in imposex, indicating further contamination (Minchin *et al.*, 1995). In Cork Harbour, a major shipping port on the Irish south coast, dogwhelk populations no longer exist on the western side of the Harbour and measurements of the condition of intersex in periwinkles (Bauer *et al.*, 1995) clearly demonstrated that this part of the Harbour was the most contaminated (Minchin *et al.*, 1996). This investigation in the Killybegs region examines the contribution of fishing vessels as a source of TBT contamination by means of two snail indicator species.

Methods

TBT exposure in marine prosobranch snails is known to produce a pathological condition, termed imposex, characterised by the superimposition of male

sex characters, normally a penis, vas deferens and/or prostate gland, on females of dioecious species. In the final stages of imposex development female dogwhelks *Nucella lapillus* are sterilized by an obstruction of the pallial oviduct. The obstruction results in an accumulation of egg capsules in the oviduct which eventually ruptures killing the snail. Dogwhelk populations from even moderately contaminated areas can thus become extinct. For the quantification of imposex intensities in natural populations different indices have been introduced, as the vas deferens sequence index (VDSI: calculated as the mean value of imposex stages in a sample), the female penis length (FPL), the relative penis size (RPSI: $(\text{mean length of the female penis})^3 / (\text{mean length of the male penis})^3 \times 100$); (Gibbs *et al.*, 1987; Oehlmann *et al.*, 1991). The VDSI and the determination of the frequency of females with a blocked vulva provide useful indications of the survival prospects of a population, and were recorded in this study. Before being examined, *N. lapillus* were narcotized using 7% MgCl_2 in distilled water for 2–4 h. The periwinkle, *Littorina littorea*, was narcotized in the same way. The presence of TBT results in alterations to the pallial oviduct and replacement of female by male organs, termed intersex and resulting in reproductive impairment. Intersex in periwinkles is a gradual transformation of the female pallial tract, which can be described by a progression of five stages

(0 to 4). Stage 0 is a normal female without any intersex characteristics. In the following two stages TBT prevents the ontogenetical infolding of the pallial oviduct from the mantle epithelium with a subsequent fusion of its free edges. Consequently in stage 1 the bursa copulatrix and in addition in stage 2 the rest of the pallial oviduct is split ventrally exposing the internal lobes. In stage 3 the pallial oviduct glands are entirely or in part supplanted by a prostate gland. In the final stage 4 a penis and sperm groove are present (Bauer *et al.*, 1995). Females in the stages 2, 3 and 4 become sterile either because the capsular material leaks into the mantle cavity (stage 2) or the glands for the formation of the egg capsules are missing (stages 3 and 4). The main parameter for the quantification of intersex intensities in populations is the intersex index (ISI: calculated as the mean value of intersex stages in a sample) which was determined according to Bauer *et al.* (1995).

Collections were made from ten stations inside Killybegs Harbour (inset of Fig. 1), four at its entrance and ten outside, close to the same time in each year (June 1994 and late May 1995) to avoid known seasonal variations in determination of indices (Stroben *et al.*, 1996). Samples from two stations (8 and 9) within the Harbour, from the high and low shore, were obtained in March 1995. A further sample series was taken from the Harbour entrance (three stations) and within the Harbour (seven stations) to confirm observations made in June 1994. Up to 55 snails of both species, where available, were collected and the catch per unit effort used to gather 50 snails was recorded to the nearest minute. Snails were examined at the University of Münster. Data collected during 1987 (4 stations) and 1993 (2 stations) were used to obtain information on longer term trends for the area.

The determination of TBT compounds in snail tissue was based largely on Stroben *et al.* (1992). The complete tissue of five snails was homogenized in stoppered tubes and an aliquot of 1 g used for further processing. 10 ml of concentrated HCl was added and after shaking for 30 min the tissues were extracted with 20 ml of hexane on an automatic shaker for 60 min. TBT was separated from DBT by washing the hexane extract with 3 ml 1 N NaOH for 10 min. The washed extracts were transferred to nitric acid and quantified by ICP-MS (Perkin Elmer Elan 5000 with an AS 91 autosampler). Internal standardization (standard addition with spiked samples) was employed and additionally standard reference materials (PACS-1 and NIES-11) were analysed. All TBT concentrations are given on a dry weight basis as Sn ($\mu\text{g TBT-Sn kg}^{-1}$ dry weight).

Sediments were obtained using a Van-veen grab. The undisturbed surface of the bioturbated mud was removed with a glass slide and placed in 400 ml cleaned glass jars. These were covered with aluminium foil and chilled.

The determination of organotin compounds in sediments was performed according to Kalbfus *et al.* (1991). To 5–100 g aliquots of the sediments 50–1000 ng tripropyltin chloride as an internal standard were added, and organotin compounds were extracted on a shaker table for three cycles with 0.05% tropolone-toluene. The extract was dried over Na_2SO_4 and proceeded to derivatization. The following step included the derivatization with 1–5 ml *n*-pentylmagnesium bromide (15 min reaction time) and the reaction quenched by adding 25 ml water + 10 ml 10% HNO_3 . The organic phase was separated and collected. The clean-up step was carried out in a clean-up column filled with florisil (with 7 g 1% deactivated silica) to which the concentrated sample was applied. The column was eluted with 100 ml hexane and the eluate concentrated to a volume of 0.1–1.0 ml. *n*-Pentyl derivatives of dipropyltin were added as an external standard and proceeded to GC/MS analysis (HP 6890A II and HP 5970A).

Results

Nucella lapillus was absent from all stations within the Harbour. At four of these stations (9, 11, 15 and 18; Fig. 1) only old worn shells remained, indicating their previous existence (Fig. 2a). Live dogwhelks were found at Port Rushin (19) near the Harbour entrance on the western side and at Carntullagh Upper (8) on the eastern side. At two stations (8, 19) and nearby Richies Bay (5), all female dogwhelks sampled had an obstructed vulva. These stations also had highest RPSI and VDSI levels (Table 1). At Lighthouse Point (6) imposex levels were again high, and juveniles and adults, sampled separately, had similar imposex levels. Values elsewhere in McSwynes and Donegal Bays were less. VDSI values for dogwhelks were obtained at 12 km from source (Table 1 and Fig. 2a).

Two distant stations had higher imposex values than stations nearer the port: Cross Harbour (21) where there is a boatslip and small boats and yachts are moored, and Teelin Harbour (24), where fishing vessels may occasionally berth.

ISI values indicated contamination was greatest within Killybegs Harbour followed by Richies Bay (Fig. 2b). In 1995, the closest periwinkle population to the port had an index of 1.6 (Table 1). ISI values were similar for 1994 and 1995. Levels near the Harbour entrance are low (0.11–0.25), yet on the south-east side of McSwynes Bay (stations 1, 2 and 3) levels were greater than this (0.31–0.36) (Table 1). At one exposed station (22) an unexpectedly high ISI value of 0.42 was obtained.

Dogwhelks sampled in previous years (1987, 1993) and again in 1994 showed general increases of the three imposex indices (Table 2). The dogwhelk populations in Carntullagh Upper and Richies Bay had become

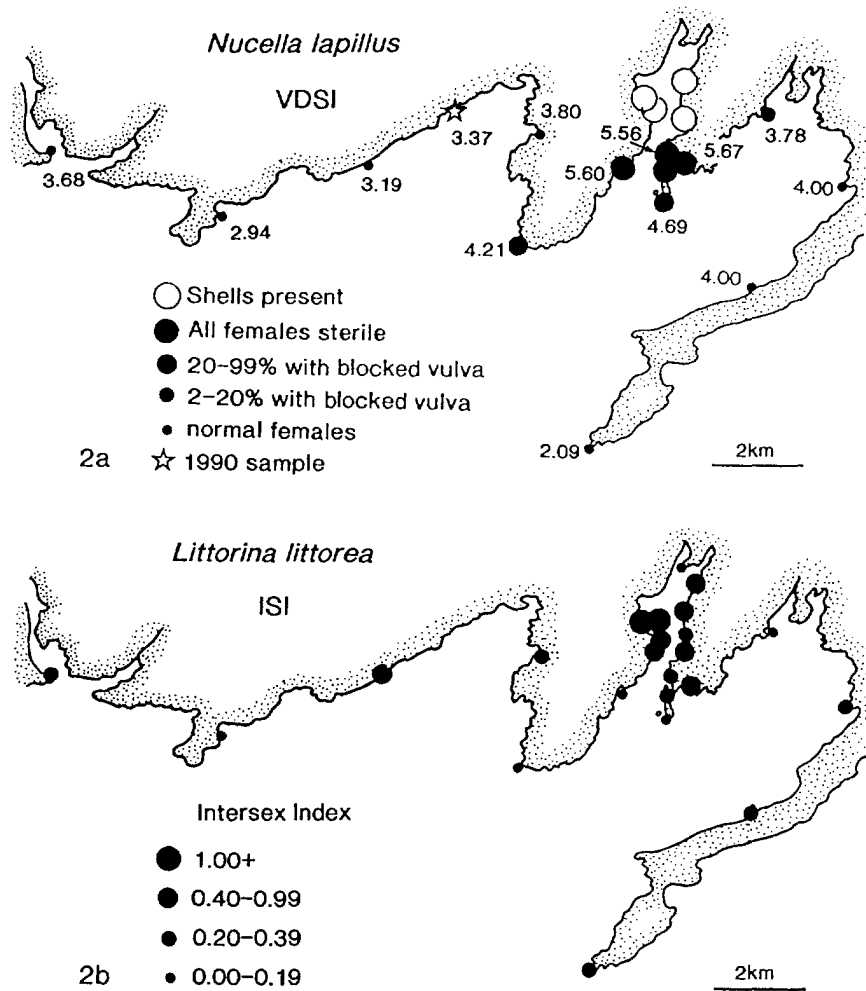


Fig. 2 (a) Imposex values and frequency of sterile dogwhelks. (b) Intersex values for periwinkles.

endangered over this time by the continual decline of females and increased reproductive impairment.

Chemical analyses of TBT body burden was consistent with the biological indices for both snails. Levels for each snail species were highest at the closest point to Killybegs and declined with straight-line distance from source (Figs 3 and 4).

Discussion

Once imposex has developed in dogwhelks the condition does not reverse itself (Bryan *et al.*, 1987; Stroben, 1994; Stroben *et al.*, 1992; Oehlmann *et al.*, 1993). For this reason it provides a good indication of past relative TBT contamination, but will not, however, necessarily reflect the current state. Evans *et al.* (1995) have questioned the general assumption that imposex is caused only by the presence of TBT. However, no further environmental compounds are likely to cause this condition in the Killybegs region apart from triphenyltin (TPT) and this is not known to have been applied in local boatyards. TPT has been recently

reported to induce imposex in some marine snails in Japan (Horiguchi *et al.*, 1995). Nevertheless imposex can be induced under selective *in vivo* conditions (Stroben *et al.*, 1992). There is little doubt that the indices found in snails in the Killybegs region reflects the presence of TBT. This substance is known to be applied at the Killybegs boatyards where many fishing vessels are repainted, has been analysed from snails at decreasing levels from source and has been identified in Harbour sediments. No other xenobiotics are known from this area. Similar patterns have been found elsewhere (Svavarsson & Skarphédinsdóttir, 1995).

The accumulations of TBT in fine sediments in the Harbour attained levels of $770\text{--}700\ \mu\text{g kg}^{-1}$ in 1995, the highest value being within 0.5 km of the source area. The breakdown products dibutyltin and monobutyltin were also present. Similar values, determined in 1993, of up to $800\ \mu\text{g kg}^{-1}$ were determined for the same region (M. O'Sullivan, pers. comm.). Although these are high values, levels five times greater, of up to $3935\ \mu\text{g kg}^{-1}$ have been found in some English estuaries (Dowson *et al.*, 1992).

TABLE 1
 Imposex and intersex values for dogwhelks and periwinkles 1994, values for 1995 in italics.

Station	Locality	<i>Nucella lapillus</i>								<i>Littorina littorea</i>					
		Sample number	VDSI	RPSI %	FPL	F %	Vulva blocked	cpue in minutes	Comment	Bodyburden g TBT-Sn kg ⁻¹	Number of females	ISI	Sterile females	cpue in mins	Bodyburden µg TBT-Sn kg ⁻¹
1	St John's Point	43	2.09	0.02	0.37	51	0%	2	E,J	84.7	25	0.36	0%	35	37.7
2	Ballysagaart Bay	52	4.00	12.34	2.94	46	0%	3	E,J	170.3	16	0.31	0%	2	118.1
3	Riggyheugh	55	4.00	5.16	2.13	61	0%	3	E,J	191.7	21	0.33	0%	3	91.6
4	North of Binroe Point	55	3.78	1.65	1.55	74	2%	28	E,J	161.9	30	0.03	0%	11	34.8
5	Richles Bay	49	5.67	35.93	4.53	12	100%	5	E,J	702.7	16	0.56	6%	1	203.1
6	Lighthouse Point	45	4.69	53.76	4.74	29	46%	5	E,J	713.7	19	0.11	0%	2	ND
6	<i>Lighthouse Point</i>	20	<i>5.00</i>	<i>42.52</i>	<i>4.80</i>	<i>10</i>	<i>100%</i>			<i>482.6</i>	<i>24</i>	<i>0.17</i>	<i>0%</i>	<i>4</i>	<i>191.5</i>
7	<i>Green Island</i>	15	<i>all males</i>			<i>0</i>				<i>101.6</i>	<i>18</i>	<i>0.22</i>	<i>0%</i>	<i>5</i>	<i>52.5</i>
8	Carntullagh Upper	48	5.56	56.16	5.24	19	100%	9	J	649.8	16	0.25	0%	3	296.5
8	<i>Carntullagh Upper</i>										22	<i>0.14</i>	<i>0%</i>		<i>120.7</i>
9	Walker's Bay	0						Absent	Shells		20	0.60	10%	3	214.3
9	<i>Walker's Bay</i>	0						<i>Absent</i>			25	<i>0.40</i>	<i>0%</i>	<i>3</i>	<i>194.3</i>
10	<i>Black Rock</i>	0						<i>Absent</i>			17	<i>0.35</i>	<i>0%</i>	<i>4</i>	<i>94.4</i>
11	Opposite Killybegs Pier	0						Absent	Shells		16	0.50	0%	2	ND
11	<i>Opposite Killybegs Pier</i>	0						<i>Absent</i>			27	<i>0.74</i>	<i>11%</i>	<i>3</i>	<i>149.6</i>
12	Opposite fish factory	0						Absent			24	0.42	0%	5	ND
12	<i>Opposite fish factory</i>	0						<i>Absent</i>			28	<i>0.39</i>	<i>0%</i>	<i>4</i>	<i>117</i>
13	Killybegs Inner Harbour	0						Absent			3	0.00	0%	12+	156
14	Main port area	0						Absent			0			absent	
15	St Catherine's Well	0						Absent	Shells		15	0.67	7%	5	209.4
15	<i>St Catherine's Well</i>	0						<i>Absent</i>			11	<i>1.00</i>	<i>18%</i>	<i>4</i>	<i>311.7</i>
16	<i>West Smooth Point</i>	0						<i>Absent</i>			15	<i>1.60</i>	<i>47%</i>	<i>35</i>	<i>502.3</i>
17	East Smooth Point	0						Absent			15	0.60	7%	17	230.1
18	Clay cliffs, Rough Island	0						Absent	Shells		16	0.44	6%	45	185
19	Port Rushin	60	5.60	31.95	4.20	8	100%	18	J	618.9	34	0.12	0%	6	182.1
20	Drumanoo Head	54	4.21	12.65	3.31	35	21%	8	E,J	216.7	28	0.07	0%	17	107.3
21	Cross Harbour	54	3.80	1.51	1.72	56	0%	8	E,J	147.3	13	0.23	0%	4	82.6
22	Kilcar coastal road fork	55	3.19	0.78	1.06	38	0%	3	E,J	107.7	12	0.42	0%	6	90.2
23	Traban, Muckcross Point	59	2.94	0.20	0.86	52	0%	3	E,J	61.3	19	0.16	0%	2	19.4
24	Teelin Harbour	55	3.68	0.85	1.60	45	0%	2	E,J	159.1	17	0.35	0%	11	118.3

F%, Percent females present; cpue, catch per unit effort—time to collect 50 snails; E, eggs; J, Juveniles

TABLE 2

Imposex levels in the dogwhelk, *Nucella lapillus*, since 1987 in the Killybegs area.

Locality	Year	Sample size	VDSI	RPSI%	FPL (mm)	%F	% with blocked vulva
Ballysagart Bay (station 2)	1987	50	ND	4.1	1.21	56	ND
	1994	52	4.00	12.3	2.94	46	0
Riggyheugh (station 3)	1987	50	ND	0.6	0.67	58	ND
	1994	55	4.00	5.2	2.13	61	0
Richies Bay (station 5)	1987	50	4.55	25.8	2.20	50	ND
	1993	27	5.00	21.7	3.73	11	67
	1994	49	5.67	35.9	4.53	8	100
Lighthouse Point (station 6)	1994	45	4.67	53.7	4.74	29	46
	1995	20	5.00	75.2	5.00	10	100
Carntullagh Upper (station 8)	1987	50	4.61	34.9	2.80	40	ND
	1993	52	5.78	64.0	5.61	17	100
	1994	48	5.56	56.2	5.24	18	100
	1995	15	—	—	—	0	No females

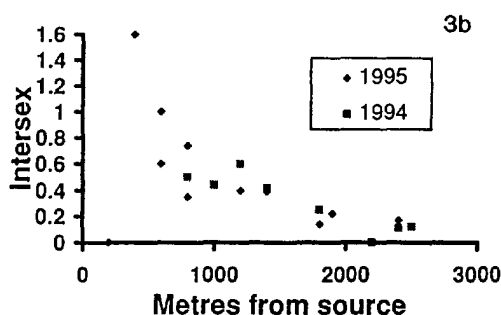
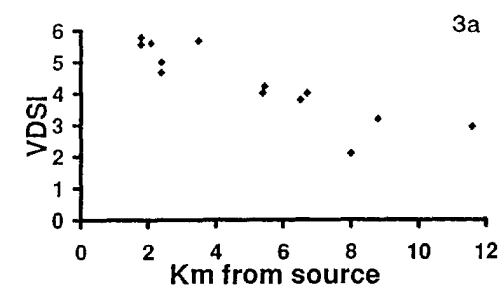


Fig. 3 (a) Imposex values with distance from Killybegs port. (b) Intersex values with distance from Killybegs port.

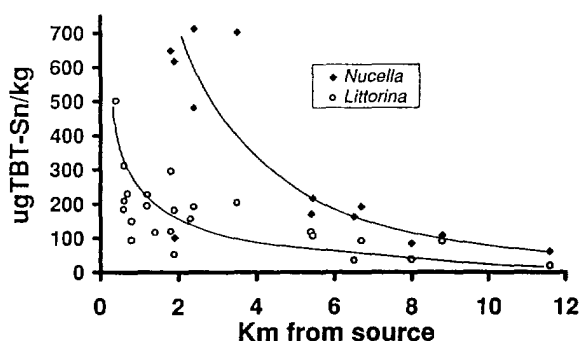


Fig. 4 TBT concentration in tissues with straight line distances by sea from Killybegs port.

It is likely that TBT and its breakdown products will persist in the Harbour sediments for some years following the time when it is no longer used in antifouling paints. The sewage discharge into the Harbour from the population of less than 5000, and fish and fish processing wastes may be important in maintaining high levels of organotin contamination because organotins have a high affinity for organic-rich sediments (Langston & Pope, 1995) and especially in the presence of humic acids (Kuballa *et al.*, 1995; Fent & Looser, 1995). Sediments collected in 1995 adjacent to stations 11 and 17 had 14.6% and 15.2% volatile suspended solids dry weight. Resuspension of sediments by ship movements, turbulence, tidal activity and dredging will redistribute TBT from this source into the water column (Dowson *et al.*, 1992) and cause it to become dispersed from the Harbour area.

TBT levels are sufficiently high inside the Harbour to have caused the disappearance of dogwhelks. By 1987, they had become extinct at Walker's Bay, as shown by their shells strewn on the upper shore. Those stations with a time series show increases in imposex levels indicating that organotin contamination within Killybegs Harbour has increased, and the disappearance of dogwhelks within the Harbour is likely to extend to the Harbour entrance. At Carntullagh Upper there was a balanced sex ratio (40% females) and an abundance of dogwhelks in 1987. By 1993 the ratio of females had become reduced to 20%, all with a blocked vulva, and levels of TBT in tissues were highest in these dogwhelks (Fig. 4). In 1995 no females were found in this area and consequently no imposex values were obtained, yet these snails had unexpectedly low burdens. It is expected that the remaining males will expire within a few years. There are changes in Richies Bay (5) and at the Lighthouse Point (6). There has been a noticeable decline of dogwhelk abundance in Richies Bay since 1987, where in 1994 all females sampled were incapable of laying eggs. However, some females in the population must have been capable of reproduction at that time

since eggs and juveniles were observed. The salmon farm near station 5, established in 1985, never used organotins and consequently the farm can not be considered as a one-time source. The population decline, sterility of female dogwhelks and notable rise in the degree of imposex since 1987, as in Carntullagh Upper, suggest a persistent and increasing level of contamination.

Contamination in Richies Bay may be higher than expected because of local water movements. It has been suggested that the high nutrient water in Killybegs Harbour may be flushed to Richies Bay, deduced from the periodically elevated levels of chlorophyll found there (Anon., 1988).

Two stations on the south-east side of McSwynes Bay showed slight increases in RPSI values since 1987, and VDSI values of 4.0 show a noticeable increase with time up to 5 km from source. Evans *et al.* (1995) suggested that RPSI values of < 25, have little effect on dogwhelk reproduction and the results in this study are in agreement.

The highest ISI indices for *L. littorea* at Killybegs attained levels similar to those found near a shipyard and ferry terminal in Cork Harbour (Minchin *et al.*, 1996). The importance of discharges from shipyards has been well established (Quevauviller *et al.*, 1989).

ISI values do not provide the same overall pattern as was found for dogwhelk VDSI, RPSI, RPLI and FPL. Although the highest values fall within the area where dogwhelks have become extinct, low levels are found at the harbour entrance (ISI range 0.11–0.22) where dogwhelk imposex would suggest that TBT levels are elevated, indicating the relative lack of sensitivity of periwinkles. In Cork Harbour it was observed that at ISI values > 0.1 some *N. lapillus* will have developed a blocked vulva, and once it exceeds 0.40 dogwhelk populations may be endangered (Minchin *et al.*, 1996). This same trend was evident in the Killybegs region although there were some differences. The higher than expected figures in the south-east region of McSwynes Bay (stations 1, 2 and 3), with an ISI range 0.31–0.36, may be due to ships that seasonally moor here for some weeks. However, imposex values for this region were low and no dogwhelks were found with a blocked vulva. The ISI value of 0.42 below the Kilcar coastal road fork is similarly difficult to explain as this is remote from areas where boats might be expected to shelter.

The lowest ISI level found within Killybegs Harbour was near the small river estuary in the inner Harbour (13). Only three female snails were found in this area high on the shore, where they would not have been covered by the tide for long periods and were close to inputs of freshwater. This suggests that increased height on the shore may result in lower degrees of intersex or that the water quality high on the shore contained low levels of organotins. To test this hypothesis snails collected from the high and low shore at Walkers Bay (ISI upper shore 0.33, lower shore 0.37) and Carntul-

lagh Upper (upper shore 0.17, lower shore 0.29) were examined and demonstrated no statistically significant difference although there was a trend of higher values lower on the shore. The values for the two stations are lower in 1995 than those obtained the previous July; these differences may not be significant but demonstrate that contamination remains at a high level. Although vertical shore movements of the periwinkles may take place, the higher parasite loadings found in those on the upper shore suggest that there is little movement between high and low water levels. ISI values at the lower end of the range are less useful for the discrimination of organotin levels and this hypothesis should be examined in an area with greater contamination. Should there be real differences in ISI values with height on the shore, this may relate to differences in submergence and/or grazing time.

The highest ISI level found in Killybegs Harbour (1.60) was considerably lower (indicating less TBT contamination) than values that may exceed 3.0 in some German ports (Bauer *et al.*, 1995). In their study a relationship between TBT contamination levels and intersex was shown. In Germany values to 3000 µg TBT Sn kg⁻¹ (dry weight) were obtained in port areas (Oehlmann *et al.*, 1996). Kure & Depledge (1994), working in Denmark, found *L. littorea* capable of maintaining a population in areas near marinas where TBT levels in tissue are up to 2080 µg TBT Sn kg⁻¹ (dry weight). The high levels at the marina site in Denmark may reflect the later 1991 Danish legislation controlling TBT.

Nucella lapillus is more sensitive to the effects of TBT contamination than *L. littorea* and comparison between intersex and imposex values is not possible since dogwhelks become extinct at ISI values of < 0.5 (< 15% of the intersex range). *Littorina littorea* is tolerant of higher organotin concentrations and naturally ranges into brackish water conditions providing a wider geographic area over which sampling can take place. As most port areas are in estuaries its higher tolerance of TBT and reduced salinity makes *L. littorea* especially useful. However, in open coastal areas, where contamination is generally lower, dogwhelks remain the more useful bioindicator, as ISI values in periwinkles are low and may be imprecise when compared to the broader ranges in VDSI and RPSI expressed in dogwhelks.

Conclusions

Nucella lapillus was a useful bioindicator for monitoring organotin contamination at up to 12 km from the main source, Killybegs Harbour, a fishing port. Its gradual population decline near the Harbour entrance and elevated imposex values indicate contamination in this region has increased. TBT in the Harbour is released by fishing boats and boatyards and is also released from the sediment where it may be stored,

possibly aided by accumulations of organic wastes. In contaminated areas where dogwhelks are not found, intersex stages in *L. littorea* allowed useful monitoring. The continuing TBT contamination in Killybegs is likely to result in further dogwhelk extinctions at stations near the Harbour entrance. Restrictions on the usage of TBT imposed in 1987 have had no effect in reducing TBT in this major fishing harbour. This is because many of the fishing vessels at Killybegs exceed 25m, and so are not subject to antifouling controls. Should restrictions on the use of TBT paint applications be increased to vessels of up to 50m as has been suggested by the International Maritime Organisation (Anon., 1994) contamination would be expected to remain in this Harbour for some years.

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