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Survey of Organotin Compounds in the Western Mediterranean Using Molluscs and Fish as Sentinel Organisms

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Abstract. Tributyltin (TBT) and its degradation products, mono- (MBT) and dibutyltin (DBT) as well as triphenyltin (TPT) were determined in molluscs and fish collected along the Catalan coast (Western Mediterranean). Marine molluscs (mussels, clams and snails) were sampled from three harbors with different characteristics (small *vs.* large boats). Two fish species were studied (a) the grey mullet *Liza aurata* sampled in Barcelona harbor and (b) the red mullet *Mullus barbatus* sampled along the coast; different tissues (muscle, liver, gills and digestive tube) were analysed separately. The composition of butyltin compounds was different according to the organism and sampling point, but in general elevated concentrations of TBT were noticed in molluscs. The highest organotin residue levels (5.4 µg/g d.w. as Sn) were detected in mussels from Masnou, a recreational marina, followed by those collected in Barcelona harbor (1.2 µg/g d.w. as Sn). In contrast, no organotin compounds were detected in fish muscle and very low levels in the other organs, being TPT the major organotin in red mullet liver.

Organotin compounds are used in a variety of consumer and industrial products including marine antifouling paints, agricultural pesticides, wood preservatives, and plastic stabilizers. It is widely accepted that antifouling paints are the most important contributors of organotin compounds to the marine environment where they have been responsible for many deleterious effects to nontarget aquatic life (WHO 1980; Alzieu 1991; Ruiz *et al.* 1995). Accordingly, several countries have banned its use and restricted its application to large vessels. However, after regulation of TBT in antifouling paints, monitoring programmes still point out the presence of organotin compounds in coastal areas (Waite *et al.* 1991; Tolosa *et al.* 1992; Dowson *et al.* 1993; Minchin *et al.* 1995; Becker-van Slooten and Tarradellas 1995). Thus, marine organisms are still subjected to exposure from sediment, water-column and through their diet, being the accumulation of TBT in certain species extremely high and rapid (Becker-van Slooten and Tarradellas 1994).

Moreover, it is reported that organotin compounds degrade slowly in the environment, with half-life times of TBT in water between one and three weeks (Seligman *et al.* 1986, 1988), being the degradation of TBT in sediment slower, with nominal half-lives on the order of 1 to 5 years (Waldock *et al.* 1987; Adelman *et al.* 1990). Therefore, the persistence and toxicity of organotin compounds makes necessary the assessment of pollution levels in biota samples, particularly in those organisms living in contact with the sediment. Marine bivalves are among the most studied organisms, they have shown a limited ability to metabolise TBT (Lee 1986), and therefore they have been chosen as sentinel organisms in pollution monitoring programmes (Laughlin *et al.* 1986a; Wade *et al.* 1988; Becker *et al.* 1992; Uhler *et al.* 1993; Kure and Depledge 1994; Stäb *et al.* 1995). However, while contamination and toxic effects of organotin compounds in lower trophic aquatic organisms is well-documented, few reports are available on the accumulation of organotin compounds by marine fishes, particularly benthic ones. The bioconcentration of organotin compounds by marine and freshwater fish has been extensively studied in the laboratory (Tsuda *et al.* 1988, 1992; Martin *et al.* 1989; Yamada *et al.* 1992, 1994), but field data are limited (Short and Thrower 1986; Krone *et al.* 1989; Kannan *et al.* 1995).

The objectives of the present study were (1) to assess the relative bioavailability of organotin compounds by looking at different species and habitats and (2) to provide data on the occurrence of organotin compounds, including mono- (MBT), di- (DBT), tri-butyltin (TBT), as well as triphenyltin (TPT) in molluscs and fishes from the Western Mediterranean (Spanish coast). The Mediterranean coastline has become an important tourist destination over the past decade, therefore an increasing number of yacht marinas have appeared. However, data on occurrence of organotin compounds in the area (water, sediment or organisms) is still scarce (Gabrielides *et al.* 1990; Tolosa *et al.* 1992) and little is known about temporal variation and behaviour of organotin compounds in this semi-enclosed body of water (Alzieu *et al.* 1991).

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Material and Methods

Sampling Sites

Organisms were collected in July 1995 from the area indicated in Figure 1. Different mollusc species (mussels, *Mytilus galloprovincialis*, clams, *Tapes decussata*, and snails, *Thais haemastoma*) were sampled in Masnou (a recreational marina), Barcelona (a commercial harbor), and St. Carles (a fishing harbor) depending on availability. In St. Carles (Ebro Delta), mussel and clam samples were also taken from the marine farms located in the bay, in order to compare levels of contamination inside and outside the harbor (Figure 1). The studied fishes were: (1) grey mullet, *Liza aurata*, collected in Barcelona harbor, and (2) red mullet, *Mullus barbatus*, sampled offshore as indicated in Figure 1. Organisms were carried dry to the laboratory within 1–2 h after collection, immediately frozen and stored at −20°C until analysis. At each sampling point, triplicate samples of a minimum of 5–6 animals pooled together were analyzed.

Organotin Analysis

Analyses were based on those described by Kuballa *et al.* (1995). Briefly, frozen samples (5–6 pooled organisms) were thawed and cut into small pieces. Dry-weight percentages were determined by freeze-drying approx. 2 g of the sample for 24 h. Subsamples (1–2 g of wet tissue) were digested with 20% tetramethylammonium hydroxide (10 ml); the mixture was kept for 2 h at 60°C. After the digestion step, the pH was adjusted to 5 with 2.0 M sodium acetate-acetic acid buffer and the following reagents added to the buffered solution subsequently: 50 ng of tetrabutyltin as internal standard, 10 ml of *n*-hexane and 4 ml of freshly made 2% sodium tetraethylborate as the ethylation agent. Afterward, the sample was shaken for 1 h, the hexane layer was collected, and the aqueous phase extracted again twice with 10 ml of *n*-hexane. The combined organic phase was reduced to approx. 1 ml., passed through a short column of 3% water deactivated alumina and eluted with 10 ml of *n*-hexane to remove lipids. The final extract was injected into a gas chromatograph (GC) with flame photometric detector (GC-FPD) and a tin mode filter (610 nm). A fused silica capillary column (30 m length × 0.25 mm i.d.; DB-17) was used for GC separation. Identification of organotin compounds was made by assigning peaks in samples to the corresponding peaks of external standard. Peak areas of individual organotin compounds were used for the quantification and results corrected for the recovery of the internal standard (94% ± 5; $n > 10$). Procedural blanks were processed with every set of samples, they were all free from organotin contamination or other interferences. The accuracy of the analytical method was checked using a certified reference biological material (NIES-11), being the recovery of TBT (71% ± 3; $n > 10$). The limits of quantification (3 times base line) were about 5 ng/g d.w. for DBT and TBT and 13 ng/g d.w. for TPT.

Results and Discussion

Residue Levels

The maximum concentration of organotin compounds was found in molluscs collected in Masnou marina (Table 1), a recreational harbor with approximately 500 moored boats. Mussels from Masnou showed a mean concentration of total organotin compounds of 5.4 ± 1.8 µg/g d.w. as Sn; which was 4.5-fold higher than the concentration found in mussels from Barcelona, a harbor with an important commercial traffic, and 44-fold higher than the residue detected in mussels from St. Carles, a fishing harbor with approx. 200 moored boats. The concentration of TBT detected in mussels from Masnou marina was 3.5 µg/g d.w. (Table 2). Above 2 µg/g d.w. of TBT, significant adverse biological effects can be measured in adult mussels (Page and Widdows 1991; Widdows and Page 1993). Clam samples were only available in Barcelona harbor and they showed a concentration of organotin compounds similar to the one detected in mussels (Table 1). The gastropod *Thais haemastoma* was sampled in Masnou and Barcelona harbors, and specimens from Masnou showed the highest concentration of organotin compounds, twofold higher than those from Barcelona.

With regard to fish analysis, the concentration of total organotin compounds was considerably lower. Grey mullet, *Liza aurata*, was sampled in Barcelona harbor and no organotin compound was detected in the muscle. The highest concentration was detected in the liver (187 ng/g d.w. as Sn) which is sixfold lower than the value reported in mussels and clams from the same area, and similar to levels determined in snails. Samples of red mullet, *Mullus barbatus*, caught offshore along the coast, showed lower concentration of organotin compounds in the liver (88–133 ng/g d.w. as Sn) than grey mullets, and surprisingly, not significant differences among sampling points were detected. The fact that no organotin compound was detected in the muscle of the studied fish points out the high metabolic potential for organotin compounds in these organisms, but also stresses the poor transference of these contaminants through the trophic chain.

Patterns of Occurrence

Molluscs: The pattern of occurrence in molluscs of TBT and its metabolites (DBT, MBT) as well as TPT is reported in Table 2. In general, TBT was present at substantially higher levels than DBT or MBT, particularly in Masnou area. This indicates that the fresh inputs of the antifouling agent dominate the degradation product. The sampling coincided with the greatest sailing activity in the area, thus probably new TBT is added to the environment. Kure and Depledge (1994) reported that during the winter, only 20–40% of organotin load was TBT in the gastropod *Littorina littorea*, but this proportion increased in spring to 60–70%.

Tributyltin/dibutyltin ratios were therefore studied in molluscs samples to obtain insight into metabolic/degradation processes or differences in availability depending on the habitat of the organism. From our limited set of data, it appears that the ratio TBT/DBT is species dependent, viz. if we look at Barcelona harbor, TBT concentrations in clams were much higher than DBT, the average TBT/DBT ratio for clams was 3.6 ± 0.9, whereas the average ratio for mussels was 0.8 ± 0.1, and snails 1.2 ± 0.8 (mean ± standard deviation). These results may indicate a certain ability of mussels and snails to metabolize TBT in contrast to clams; or more likely, a different availability of butyltin species for clams, as they live buried in sandy substrates. Alternatively, looking at mussel samples from all three locations, TBT/DBT ratio increased with TBT concentration ($r = 0.76$). This observation can be due to differences in exposure; samples with high TBT/DBT ratios are near sources and at these locations, water concentrations of TBT are probably high compared to metabolites and this is reflected in

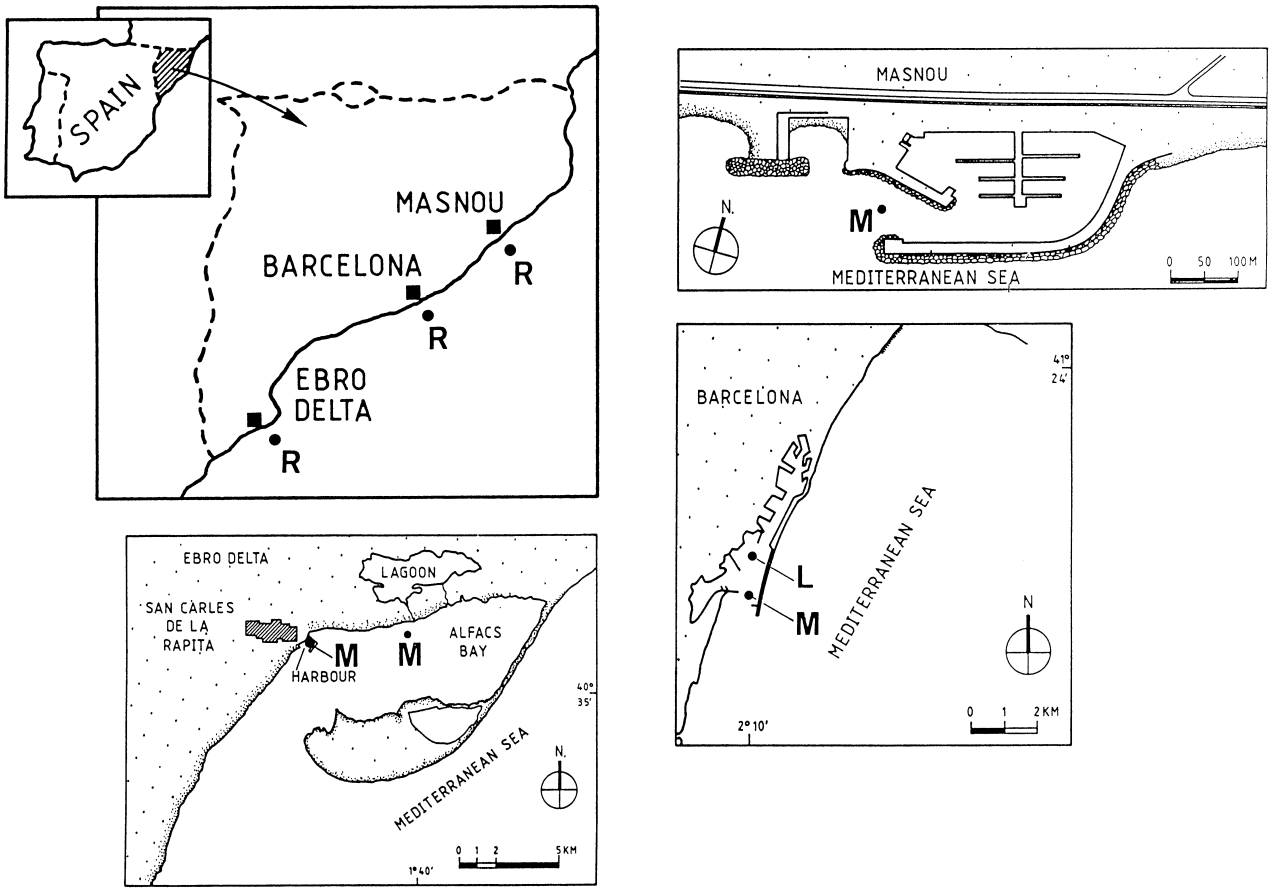

Fig. 1. Map showing sampling areas and stations: (M) mollusc samples, (L) grey mullet *Liza aurata*, (R) red mullet *Mullus barbatus* sampled offshore

Table 1. Concentration of total organotin compounds (ng/g d.w. as Sn) in molluscs and fish sampled along the Catalan coast (Western Mediterranean). n.d. = not detected

Organism	Masnou	Barcelona	St. Carles
Mytilus galloprov.	5444 ± 1816	1165 ± 18	123 ± 17
Tapes decusata	—	1094 ± 156	—
T. haemastoma	506 ± 305	232 ± 84	—
L. aurata			
muscle		n.d.	
liver		187 ± 6	
gills		36 ± 7	
M. barbatus			
muscle	n.d.	n.d.	n.d.
liver	113 ± 34	133 ± 28	88 ± 25
gills	56 ± 4	169 ± 12	89 ± 4
dig. tube	77 ± 7	161 ± 9	78 ± 4

— = Sample not available.

the organism. Similar results are reported for zebra mussels from 56 locations in the Netherlands by Stäb *et al.* (1995).

Triphenyltin was only detected in Masnou samples (Table 2). This compound is also highly toxic to aquatic organisms and it has been used as a co-toxicant with TBT in some long-performance antifouling paints (Fent and Hunn 1991). The first report of TPT in Mediterranean coastal samples was published by Tolosa *et al.* (1992) indicating highest concentration in marina samples, which infers that contamination probably arises from the antifouling of small vessels. Diphenyltin, which is most likely the primary degradation product of TPT was not found.

Looking at the molluscs results, the contamination by organotin compounds was much lower in St. Carles harbor than in the other studied areas, but nevertheless detectable. However, mussel and clams taken from the marine farms (Alfacs bay) located approx. 3 kilometers away from the harbor (Figure 1) showed very low or even undetectable levels of TBT (Table 2). Dissipation of TBT occurs over several kilometers from the major point source, and this was previously described by Tolosa *et al.* (1992) in water samples; in addition, the low traffic of boats in the bay and the continuous fresh-water flushing from land to the sea may prevent these compounds to accumulate in the area.

Fish: Table 3 shows the pattern of occurrence of TPT, TBT and its metabolites in fish samples. Unlike molluscs, where TBT was found in the highest concentration, TPT was the predominant organotin measured in red mullet liver. Concentrations of TBT in the liver ranged from non detected to 27 ng/g d.w., whereas concentrations of TPT ranged between 77 and 112 ng/g d.w. as Sn, depending on sampling site. Among the tissues and

Table 2. Butyltin and phenyltin residues (ng/g d.w. as Sn) in molluscs sampled in three harbors along the Catalan coast (Western Mediterranean). n.d. = not detected

Organism	MBT	DBT	TBT	TPT
Masnou				
Mytilus galloprov.	115 ± 2	1506 ± 422	3516 ± 1283	311 ± 117
Tapes decusata	—	—	—	—
T. haemastoma	n.d.	104 ± 66	158 ± 97	243 ± 142
Barcelona				
Mytilus galloprov.	101 ± 5	602 ± 35	461 ± 22	n.d.
Tapes decusata	95 ± 31	238 ± 57	761 ± 73	n.d.
T. haemastoma	10 ± 10	99 ± 7	98 ± 27	n.d.
St. Carles				
Mytilus galloprov.	16 ± 16	44 ± 4	67 ± 3	n.d.
Marine farms				
Mytilus galloprov.	n.d.	n.d.	12 ± 1	n.d.
Tapes decusata	n.d.	n.d.	n.d.	n.d.

— = Sample not available

Table 3. Butyltin and phenyltin residues (ng/g d.w. as Sn) in different tissues of grey mullet *Liza aurata* sampled in Barcelona harbor and red mullet *Mullus barbatus* sampled offshore along the Catalan coast (Western Mediterranean). n.d. = not detected

Organism	Liver	Gills	Digestive Tube
Mullus barbatus			
Masnou			
MBT	n.d.	n.d.	n.d.
DBT	n.d.	n.d.	n.d.
TBT	n.d.	25 ± 3	28 ± 12
TPT	112 ± 35	31 ± 3	45 ± 7
Barcelona			
MBT	n.d.	n.d.	n.d.
DBT	7 ± 1	10 ± 1	19 ± 3
TBT	27 ± 13	110 ± 7	85 ± 2
TPT	99 ± 16	49 ± 4	57 ± 8
St. Carles			
MBT	n.d.	n.d.	n.d.
DBT	5 ± 5	7 ± 1	28 ± 5
TBT	6 ± 6	38 ± 3	90 ± 6
TPT	77 ± 24	45 ± 2	78 ± 2
Liza aurata			
Barcelona harbour			
MBT	36 ± 6	n.d.	—
DBT	74 ± 12	5 ± 2	—
TBT	78 ± 9	2 ± 2	—
TPT	n.d.	n.d.	—

— = Sample not available.

organs examined, the concentration of TPT was highest in the liver, and decreased in the order liver ≫ gills > digestive tube. On the contrary, TBT concentration was highest in gills, and decreased in the order gills > digestive tube ≫ liver. Moreover, no organotin residue was detected in muscle, probably due to the ability of fish to metabolize TBT, which enters either via food or water (Tsuda *et al.* 1988; Martin *et al.* 1989; Lee 1991). These results are consistent with exposure experiments carried out by Tsuda *et al.* (1991, 1992) and Yamada *et al.* (1992).

Generally, patterns and concentrations of hydrophobic pollutants in aquatic organisms are determined not only by their concentrations in external environmental compartments such as water, sediment and food, but also by internal physiological processes, such as lipid metabolism and biotransformation (Barron 1990). In the present study, the distribution pattern of TPT in *M. barbatus* was different from that of TBT (Figure 2). These results suggest that the metabolic pathways and activities of these two organotin compounds are different, and that TBT might be easily adsorbed on the gills in contact with seawater or ingested via food and rapidly metabolized in the liver where TBT only represents 8.9% of the total organic tin detected. The uptake of TPT via water or food appears to be similar than the uptake of TBT as shown in Table 3, nevertheless TPT represents 87.6% of the total organotin detected in the liver. These results can be interpreted as an indication that TPT is subjected to a lower rate of metabolization than TBT in red mullet, which is supported by the fact that no degradation products (DPT, MPT) were detected. In general, the tendency of a toxicant to accumulate in various tissues is highly dependent on its lipophilicity. Since the log octanol/water partition coefficient (kow) has been recorded to be in the range between 3.2 and 3.5 for TBT (Maguire *et al.* 1983; Laughlin *et al.* 1986b) and 2.1 for TPT (Tsuda *et al.* 1986), the highest concentration of both contaminants would be expected in the liver, the organ with the highest lipid content. However, trialkyltin compounds are known to bind to aminoacids, peptides and proteins (Davies and Smith 1980), and this complexation between the trialkyltin compounds and protein may also influence the tissue distribution. Yamada *et al.* (1992) observed that the accumulation of both TBT and TPT was not related with the lipid content in the tissues and organs of several marine fish species.

This is the first report on the contamination of fish by TPT in the Mediterranean area. The tendency of TPT to accumulate in marine organisms was first noticed by Takami *et al.* (1988); the authors found that although the annual production of TPT in Japan was about one tenth that of TBT, TPT residues in biota samples were higher than TBT. Thus, given its toxicity (Brüschweiler *et al.* 1995) and tendency to accumulate in biota, further efforts should be addressed to determine the extension of this contamination. The origin of TPT in red mullet samples is unknown. The results indicate that its use as an antifouling agent appears to be restricted to Masnou Marina, therefore its presence offshore could be associated to its use as a fungicide

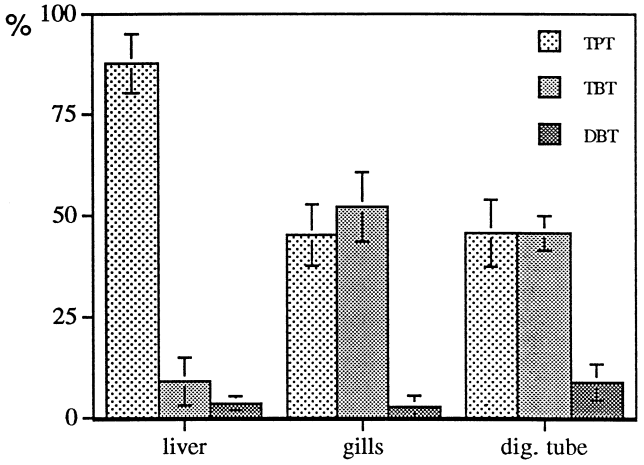

Fig. 2. Distribution of triphenyltin (TPT) and tri- and di-butyltin (TBT, DBT) in different tissues of red mullet sampled along the western Mediterranean coast

and possibly, atmospheric transport to the sea. This assumption is supported by the fact that some modelling studies carried out by Baart and Diederen (1991) estimated that volatilization is an important transport route for TPT and that TPT used in potato crop fields in the Netherlands has been detected in rainwater at remote locations (Stäb *et al.* 1994). The presence of TPT in other fish species from the Mediterranean coast or in marine sediments should be investigated in the future. It should be considered that red mullets are benthic species feeding on small organisms inhabiting the sediment, and sediments are reported to be important environmental sinks for TBT in the marine environment and an important route of uptake (Langston and Burt 1991).

For comparison, samples of grey mullet-*Liza aurata*-were taken in Barcelona harbor and muscle, liver and gills analyzed (Table 3). The ability of both grey mullet and red mullet to metabolize TBT is clearly shown by the ratio TBT/DBT, which varied from 5.7 in gills (uptake) to 1.1 in liver of grey mullet, and from 5.2 in gills to 2.5 in liver of red mullet. These results confirm the role of the liver in the metabolism of TBT as indicated by other authors (Martin *et al.* 1989). On the other hand, the concentration of butyltin compounds in the liver were between 5- to 18-fold higher in grey mullet than in red mullet liver, which is in accordance to the fact that they live closest to the potential sources. However, no TPT residue could be detected in grey mullet liver.

In summary, the present study reports on the significant accumulation of TPT and body distribution of organotin compounds in fishes from the Mediterranean coast. However, the significance of these findings for the organism health and the extention of the contamination to other species and areas should be further investigated.

Regional Comparison

The concentration of butyltins reported in molluscs in the present study are in the range of those detected in other areas, viz. in the Mussel Watch program carried out in the United States, the concentration of total butyltin reported for mussels and oysters varied from <0.005 to 3.76 µg/g Sn d.w. (Wade *et al.* 1988; Uhler 1993). The low values are from ''clean areas'' far-away from sources, and they are similar to the ones found in samples from the marine farms located in the bay (Ebro Delta); whereas the high values are in the range of those detected in Masnou mussels (5.4 µg/g d.w.). Similarly, a large variability on the concentration of organotin compounds has been reported for mussels collected from marine (Langston *et al.* 1987; Page 1995; Stäb *et al.* 1995) or fresh-water systems (Becker-van Slooten *et al.* 1992; Becker-van Slooten and Tarradellas 1995; Fent and Hunn 1995). In general, the amount of TBT detected in molluscs varies largely in function of the local sources and inputs of the compound; therefore, the analysis of these samples provides information on the amount of compound to which the organism has been exposed and that it is bioavailable.

Conversely, fish are not good sentinel organisms in organotin pollution monitoring. A regional comparison of butyltin residues in fish collected from several locations in Australia revealed comparable concentrations along the studied area (Kannan *et al.* 1995). Moreover, it has been reported that the concentration of TBT in the liver is not representative of the levels of exposure (Schwaiger *et al.* 1992).

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