The accumulation and impact of organotins on marine mammals, seabirds and fish for human consumption

A Report for WWF-UK
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Contents

1 Introduction

Organotins have been thoroughly investigated with their acute and chronic toxicity being demonstrated in a range of invertebrate marine organisms including crustaceans, polychaetes, bivalves and gastropods. Less studied is the ecotoxicological impact on fish, birds, seals and cetaceans and other marine mammals.

Organotins have also been demonstrated to have endocrine disrupting properties in fish, birds and mammals.

Organotins have been detected in a range of edible marine species, including many invertebrates such as molluscs, crustaceans and cephalopods. As organotins are bioaccumulative in certain marine species, some of which are food species used by humans, there is justifiable concern over the level of exposure of humans to organotins consumed in edible marine species.

This report will review the impact of organotins on marine mammals, fish and seabirds and the intake of organotins by humans in consuming marine food contaminated by organotins.

2 Organotins in Marine Vertebrates

2.1 Marine Mammals

There is no doubt that organotins cause harm to marine mammals. High doses of organotins have been shown to damage the central nervous system and reproductive mechanisms in mammals. The most widely used organotin, tributyltin (TBT), is an endocrine disrupting chemical in mammals. Organotins have been detected in marine mammals from all over the globe[1].

It is possible that organotins have been in part responsible for mass mortalities of dolphins found washed up in recent years.[2] Concentrations in bottlenose dolphin (*Tursiops truncatus*) liver from the US Atlantic and Gulf Coasts found between 1989 and 1994 ranged from 110 to 11340 ng/g wet weight with a mean value of 1400ng/g wet weight. These are higher than reported elsewhere. Levels in pygmy sperm whale (*Kogia briveceps*) and Atlantic spotted dolphins (*Stenella frontalis*) were also measured although these species were not affected by the large scale mortalities that lead to the dolphin carcasses being found in large numbers on the US coast. It is clear from these samples that butyltins tend to concentrate in the liver of marine mammals[3].

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1 IVM (1998) **Butyltin and phenyltin compounds in liver and blubber samples of sperm, whales (Physeter macrocephalus) stranded in the Netherlands and Denmark.** Amsterdam. IVM.

2 Kannan K, K Senthilkumar, B G Loganathan, S Takahaso, D K Odell and S Tanabe (1997) **Elevated accumulation of TBT and its breakdown products in bottlenose dolphins found stranded along the US Atlantic and Gulf coasts**. Environment. Sci. Tech. 31, 296-301

3 Kannan K, K Senthilkumar, B G Loganathan, S Takahaso, D K Odell and S Tanabe (1997) **Elevated accumulation of TBT and its breakdown products in bottlenose dolphins found stranded along the US Atlantic and Gulf coasts**. Environment. Sci. Tech. 31, 296-301

Table 1: Concentrations of butyltin compounds (ng/g wet weight) in cetaceans collected from the US Atlantic and Gulf Coasts between 1989-1994[4]

Bottlenose dolphin	**Total butyltins (with range) ng/g**
Liver	1400 (110-11340)
Kidney	200 (25-670)
Muscle	41 (13-110)
Blubber	630
Melon	190
Heart	50
Brain	110
Atlantic Spotted Dolphin	
Liver	360 (83-630)
Kidney	32 (31-33)
Muscle	21 (14-28)
Blubber	200
Pygmy Sperm Whale	
Liver	390 (350-410)
Kidney	62 (59-65)
Muscle	21 (16-26)

Concentrations of organotins have also been determined in the liver and blubber of bottlenose dolphins (*Tursiops truncatus*) found on Italy's Adriatic coast. Butyltin concentrations of 1200 - 2200 ng/g wet weight were found in liver tissue, an order of magnitude higher than in blubber[5].

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4 Kannan K, K Senthilkumar, B G Loganathan, S Takahaso, D K Odell and S Tanabe (1997) **Elevated accumulation of TBT and its breakdown products in bottlenose dolphins found stranded along the US Atlantic and Gulf coasts**. Environment. Sci. Tech. 31, 296-301

5 Kannan K, S Corsolini, S Focardi, S Tanabe, and R Tatsukawa (1996) **Accumulation pattern of butyltin compounds in dolphin, tuna and shark collected from the Italian coastal waters**. Arch Environ Contam Toxicol 31, 19-23.

Table 2: Concentrations of butyltin compounds (ng/g wet weight) in adult male bottlenose dolphins from the Italian coast of the Mediterranean Sea[6]

Tissue	Carcass Decomposition	Total butyltins (ng/g)
Blubber	Moderate	110
Liver	Moderate	1200
Blubber	Moderate	48
Blubber	Fresh	320
Liver	Fresh	2200

Concentrations of organotin compounds have been determined in liver samples from harbour porpoise (*Phocoena phocoena*) collected from the Turkish coastal waters of the Black Sea. These were in the range of 89-219 ng/g on a wet weight basis. Dibutyltin (DBT) residues were higher than those of TBT, suggesting the degradation of TBT to DBT occurs in the liver at levels comparable to other marine mammals. The results indicate that the Black Sea is also contaminated with organotin compounds, but to a lesser degree than other coastal waters such as the Mediterranean or US Atlantic and Gulf coasts. The biomagnification factor in harbour porpoises in the Black Sea was 0.8, which is comparable with pinnipeds and lower than most cetaceans[7].

Concentrations of butyltins in the liver of Risso's dolphins (*Grampus griseus*) collected off Taiji, Japan, have also been determined. Mean and range concentrations of these compounds were 3600 ng/g (550-6000 ng/g) wet weight. No difference was observed in concentrations between male and female. Concentrations increased until maturity (8-10 years) and then remained constant. Risso's dolphins showed higher biomagnification factor (about 6) than Steller sea lion (*Eumetopias ursinus*) (0.6), implying a slower excretion rate in cetaceans than pinnipeds. This is due to their lower capacity to degrade butyltins and the lack of physiological processes such as shedding of hair which removes butyltins from the body[8].

Indeed when the specific distribution of butyltins in various tissues and organs of Steller sea lions was studied in samples collected from coastal waters of Hokkaido, Japan, concentrations were an order of magnitude higher in the liver than in other tissues and organs, except hair. The levels of butyltins in hair (1500 ng/g wet weight) were the highest of all the tissues analysed. These results suggested that 25% of the total butyltin burden in the body was eliminated through shedding. Selective accumulation in liver and hair is attributed to the protein-binding capacity of butyltins rather than lipophilicity[9].

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6 Kannan K, S Corsolini, S Focardi, S Tanabe, and R Tatsukawa (1996) **Accumulation pattern of butyltin compounds in dolphin, tuna and shark collected from the Italian coastal waters.** Arch Environ Contam Toxicol 31, 19-23.

7 Madhusree B, S Tanabe, A A Ozturk, R Tatsukawa, N Miyazaki, E Ozdamar, O Arial, O Samson, and B Ozturk (1997) **Contamination by butyltin compounds in harbour porpoise (*Phocoena phocoena*) from the Black Sea**. Fresenius Journal of Analytical Chemistry. 1997; 359:244-248.

8 Kim G B, S Tanabe, R Iwakiri, R Tatsukawa, M Amano, N Miyazaki and H Tanaka (1996) **Accumulation of butyltin compounds in Rissos dolphin (*Grampus griseus*) from the Pacific coast of Japan - Comparison with organochlorine residue patterns**. Environmental Science & Technology. 1996; 30(8):2620-2625

9 Kim G B, J S Lee, S Tanabe, H Iwata, R Tatsukawa and K Shimazaki (1996) **Specific accumulation and distribution of butyltin compounds in various organs and tissues of the Steller sea lion (*Eumetopias jubatus*) - Comparison with organochlorine accumulation patterns.** Marine Pollution Bulletin. 1996;

Studies have been conducted to examine sex difference, age, and temporal trends of butyltin accumulation and its biomagnification in Steller sea lion collected from Alaska, USA, during 1976-1985 and from Hokkaido, Japan, during 1994-1995. Average concentration of butyltins in the liver of Steller sea lion from Alaska (19 ng/g wet weight) was much lower than those from western and eastern Hokkaido, Japan (150 and 220 ng/g respectively). This result suggests that Japanese coastal waters are more contaminated with butyltins than those of Alaska. In most samples, DBT residues were retained at higher levels than TBT, again suggesting the degradation of TBT to DBT in the liver. As with Risso's dolphin no sex difference in accumulation of butyltin residues was found in Steller sea lion. Unlike the dolphin, there was no age-dependent accumulation in Steller sea lion.

Similarly, no prominent temporal trend in concentrations was observed between 1976 and 1985. Nevertheless, the annual consumption of organotins doubled in the United States during the same period. These results suggest that butyltin compounds are degraded faster than the intake from diet in Steller sea lion, allowing a lower than otherwise expected and constant body burden. The biomagnification factor of butyltins in Steller sea lion was low (0.15-4.6; mean, 0.6), indicating that this animal is unlikely to magnify butyltins due to rapid degradation and excretion[10]. Cetaceans on the other hand are clearly susceptible to biomagnification and age-dependent accumulation of butyltins.

Table 3: Biomagnification factors for butyltins in marine mammals (various studies)

Steller sea lion[11]	0.6 (0.15 - 4.6)
Risso's dolphin[12]	c. 6
Harbour porpoise[13]	0.8

In UK waters, studies of butyltins in liver tissue from a range of pelagic cetaceans (such as long-finned pilot whale and minke whale), porpoises and seals have shown that butyltin contamination is also widespread in marine mammals around the UK coast. Liver tissues concentrations in pelagic cetaceans are between 19-321 ng/g, in porpoise between 22-640 ng/g and in grey seal (*Halichoerus grypus*)

32(7):558-563.

10 Kim G B, S Tanabe, R Tatsukawa, T R Loughlin and K Shimazaki (1996) **Characteristics of butyltin accumulation and its biomagnification in Steller sea lion (*Eumetopias jubatus*).** Environmental Toxicology & Chemistry. 1996; 15(11):2043-2048

11 Kim G B, S Tanabe, R Tatsukawa, T R Loughlin and K Shimazaki (1996) **Characteristics of butyltin accumulation and its biomagnification in Steller sea lion (*Eumetopias jubatus*).** Environmental Toxicology & Chemistry. 1996; 15(11):2043-2048

12 Kim G B, S Tanabe, R Iwakiri, R Tatsukawa, M Amano, N Miyazaki and H Tanaka (1996) **Accumulation of butyltin compounds in Rissos dolphin (*Grampus griseus*) from the Pacific coast of Japan - Comparison with organochlorine residue patterns.** Environmental Science & Technology. 1996; 30(8):2620-2625

13 Madhusree B, S Tanabe, A A Ozturk, R Tatsukawa, N Miyazaki, E Ozdamar, O Arial, O Samson, and B Ozturk (1997) **Contamination by butyltin compounds in harbour porpoise (*Phocoena phocoena*) from the Black Sea**. Fresenius Journal of Analytical Chemistry. 1997; 359:244-248.

between <12-22 ng/g[14]. In late 1997, male sperm whales (*Physeter macrocephalus*) beached on the coasts of Netherlands and Denmark, in the North Sea, were found with high organotin levels up to 76ng/g wet weight in the liver[15].

Table 4: Organotin levels in liver samples of stranded sperm whales stranded in 1997 (ng/g wet weight)[16]

	Netherlands	Denmark
Monobutyltin (MBT)	7	8
DBT	29	61
TBT	5	7
Total butyltins	42	76

Sea otters too have also been found contaminated with organotins. TBT and its degradation products, MBT and DBT have been determined in river, kidney, and brain tissues of adult Southern sea otters (*Enhydra lutris nereis*) found dead along the coast of California during 1992-1996. Hepatic concentrations of butyltin compounds ranged from 40 to 9200 ng/g wet wt. Concentrations in sea otters were comparable to those reported in stranded bottlenose dolphins from the U.S. Atlantic Coast during 1989-1994. Greater accumulation of organotins in sea otters may be due to their bottom-feeding habit and the diet that consists exclusively of invertebrates such as molluscs and gastropods. Livers of female sea otters contained approximately 2-fold greater concentrations of organotins than did those of males. The composition of butyltin compounds in sea otter tissues (predominantly TBT) is suggestive of recent exposure. It appears that large harbours such as Monterey that handle ships legally painted with TBT-containing antifouling paints continue to experience ecotoxicologically significant organotin contamination. Sea otters found to be affected by infectious diseases contained greater concentrations of organotins in their tissues than those found that had died from trauma and other unknown causes[17].

In 1998, Southern sea otters in the USA underwent a third consecutive year of declines, with more otters being washed up dead. Sea otters may spend considerable periods of time feeding near harbours and are therefore vulnerable to organotin contamination. It is thought that organotins have lead to immunosuppression in the sea otters making them vulnerable to infection and hence increasing mortality[18].

In conclusion, numerous marine mammals belonging to a range of species have been found to be contaminated with organotins. Butyltin compounds including TBT, DBT and MBT have been detected in almost all liver samples. Nearly all marine mammals analysed have contained butyltins. This illustrates

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14 Law R J, S J Blake, B R Jones and E Rogan (1998) **Organotin compounds in liver tissue of harbour porpoises and grey seals from the coastal waters of England and Wales.** Mar Poll Bull 36, 241-247

15 IVM (1998) **Butyltin and phenyltin compounds in liver and blubber samples of sperm, whales (*Physeter macrocephalus)* stranded in the Netherlands and Denmark**. Amsterdam. IVM.

16 **IVM (1998) Butyltin and phenyltin compounds in liver and blubber samples of sperm, whales (*Physeter macrocephalus)* stranded in the Netherlands and Denmark**. Amsterdam. IVM.

17 Kannan K, S Guruge, N Thomas, S Tanabe and J Giesy (1998) **Butyltin residues in Southern sea otters (*Enhydra lutris nereis*) found dead along California Coastal Waters** Environ Sci Technol 1998, 32 1169-1175

18 Henderson S (1998) **Sea Otter Decline** Mar Poll Bull Vol 36 No 8 p 565 August 1998

the worldwide distribution of organotins in the oceans. A notable exception is a minke whale sample from the Antarctic Ocean in which no organotins could be detected, indicating that this ocean may be relatively uncontaminated to date[19]. The elevated residues detected in coastal species and low concentrations found in off-shore species indicate a high degree of butyltin contamination in coastal waters. Mammals inhabiting waters of developed nations are, in general, found to contain higher concentrations compared with those collected from the waters of developing countries. Some mammals such as the sea lion have the ability to degrade or expel butyltins for the body, while others, such as dolphins exhibit age-dependent biomagnification of butyltins.

Due to the known endocrine disrupting ability of butyltins it is likely that negative effects are occurring in marine mammals.

Although not all marine mammals are utilised for human food, a number of different seals, whales and to a lesser extent dolphins are consumed by humans, particularly, although not exclusively, in artisanal and subsistence fishing communities. In some populations, such as the Inuit, marine mammals provide a very significant proportion of the protein and fat intake. It is likely that organotin contamination is being passed to humans consuming marine mammals as a significant part of their diet

19 Iwata H, S Tanabe, N Miyazaki and R Tatsukawa (1994) **Detection of butyltin compounds residues in the blubber of marine mammals** Mar Poll Bull 28(10) 607-612

Table 5: Butyltin compounds in liver samples of marine mammals (various studies) (ng/g wet weight)

Family	Species	Location	Sum butyltins
Physeteridae	Grt sperm whale	North Sea	42
	Grt sperm whale	North Sea	76
	Dwarf sperm whale	East Japan	730
	Pygmy sperm whale	West Japan	230
	Pygmy sperm whale	Atlantic	350-410
	Pygmy sperm whale	UK, Welsh coast	85
Globicephalidae	Grt killer whale	S-E Japan	2200-2700
	Short-finned pilot whale	East Japan	1500-2600
	Long-finned pilot whale	UK North Sea	22
Balaenopteridae	Fin whale	UK North Sea	19
	Minke whale	UK North Sea	56
Ziphiidae	Stejneger's beaked whale	W Japan	400
	Ginkgo-toothed beaked whale	W Japan	330
	Baird's beaked whale	E Japan	110-310
	Sowerby's beaked whale	UK North Sea	58
	Blainville's beaked whale	UK Welsh coast	33
	True's beaked whale	UK Irish Sea	28
Delphinidae	Bottlenose dolphin	S-E Japan	2600-3000
	Bottlenose dolphin	North Adriatic	1200-2200
	Bottlenose dolphin	Atlantic / Gulf	110-11000
	Bottlenose dolphin	Bay of Bengal	53-170
	Risso's dolphin	S-E Japan	550-6000
	Risso's dolphin	West coast, UK	66, 81
	Spinner's dolphin	Sulu Sea, Philippines	42-67
	Spinner's dolphin	Bay of Bengal	66-130
	Atlantic spotted dolphin	Florida Atlantic	83-630
	Fraser's dolphin	Sulu Sea, Philippines	89-98
	Common dolphin	UK Welsh coast	185, 263
	White-sided dolphin	UK Welsh coast	29
	White-beaked dolphin	UK North Sea	139, 170
	Striped dolphin	UK	161, 312
Stenidae	Rough-toothed dolphin	S-E Japan	3300
	Rough-toothed dolphin	West Pacific	17-37
	Humpbacked whale	Bay of Bengal	67-200
Phocoenidae	Finless porpoise	E Japan	1100
	Finless porpoise	Seto inland sea, Japan	10000
	Finless porpoise	E Japan	3300
	Finless porpoise	S Japan	5200
	Finless porpoise	China	730-1200
	Finless porpoise	China	350-430
	Dall's porpoise	Aleutians	120
	Dall's porpoise	Bering Sea	63-110

	Dall's porpoise	Kamchatka, Pacific	41-180
	Dall's porpoise	N-E Japan	340-1000
	Harbour porpoise	Black Sea	89-219
	Harbour porpoise (neonate)	Polish Baltic Coast	18-27
	Harbour porpoise	UK	22-640
Seals and sea-lions	Grey seal	UK	<12 - 22
	Larga seal	Japan	50
	Larga seal	Japan	330
	Ribbon seal	Japan	75
	Northern fur seal	N-E Japan	320
	Steller sea lion	Alaska	2-24
	Steller sea lion	Japan	75-250
	Steller sea lion	Japan	170-300

2.2 Seabirds

There is less data on organotin residues and their impact in seabirds than for marine mammals.

Laboratory studies on TBT have shown it to be embryotoxic, reducing hatching success and fertility[20]. TBT also reduces enzyme and hormone activities in Japanese quail at levels of only a few hundred ug/g[21]. However, seabirds may be less susceptible to build up very high levels of butyltins as they appear to be able to metabolise butyltins and to shed butyltins through moulting[22].

Nevertheless, in 1989 levels of butyltins in birds from Gdansk Bay were found to be very high up to 4600 ng/g in the liver of long-tailed ducks (*Clangula hyemalis*). This suggests that despite relatively high elimination and metabolism of butyltins by birds, sufficient exposure is occurring in regions such as the Southern Baltic to maintain high body burdens of butyltins[23].

Table 6: Concentrations (and ranges) of total butyltins in the liver of fish eating birds collected from Gdansk Bay in the Southern Baltic Sea in 1989

Species	Total butyltins ng/g wet weight
Red-throated diver	610
Razorbill	330 (260-380)
Great crested grebe	540
Black cormorant	870
Long-tailed duck	4600
Long tailed duck	280
Guillemot	500 (430-590)

For example, butyltin has been found in seaducks in US coastal locations at up to 1100ng/g wet weight. In the US, mollusc-feeding birds appeared to be exposed to greater concentrations of butyltins than other predatory birds feeding on fish, other birds or mammals. Exposure is also likely to be greatest in coastal bird populations feeding near harbours, marinas and ports, although the ecotoxicological significance of this is not known[24].

Liver and kidney samples from seabirds from the North Pacific, Japan and Korea have shown butyltins up to 300 ng/g wet weight in kidney and 280 ng/g wet weight in liver from cormorants (*Phalacrocorax*

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20 Schlatterer B, T M M Coenon, E Ebert, R Grau, V Hilbig, and R Munk (1993) **Effect of bis tri-n-butyltin oxide in Japanese quail exposed during egg laying period: an interlaboratory comparison study.** Arch Environ Contam Toxicol 24, 440-448

21 Coenon T M M, A Brouwer, I C Enninga and J H Koeman (1992) **Subchronic toxicity and reproduction effects of tri-n-butyltin oxide in Japanese quail.** Arch Environ Contam Toxicol 23, 457-463

22 Guruge K S, S Tanabe, H Iwata, R Tatsukawa and S Yanmagishi (1996) **Distribution, biomagnification and elimination of butyltin compound residues in common cormorants (Phalacrocorax carbo) from Lake Biwa, Japan.** Arch Environ Contam Toxicol 31, 210-217

23 Kannan K and J Falandysz (1997) **Butyltin residues in sediment, fish, fish-eating birds, harbour porpoise and human tissues for Polish Coast of the Baltic Sea**. Mar Poll Bull vol. 34 no 3 pp 203-207

24 Kannan K, K Senthilkumar, J E Elliott, L A Feyk and J P Giesy (1998**) Occurrence of butyltin compounds in tissues of water birds and seaducks from the USA and Canada** Arch Environ Contam Toxicol 35, 64-69 (1998)

carbo) (coastal birds) and 43ng/g wet weight in Laysan albatross (open ocean)[25].

Butyltins have also been found in migratory and resident birds in South India and in the Philippines. The results illustrate well avian ability to purge butyltins for the body by moulting and shedding feathers[26] [27].

Table 7: Concentration ranges in soft tissue and feathers of migratory and resident birds from India and the Philippines

	Total butyltins	
India	Soft tissue	Feathers
Migratory	<8 - 28	<9.9 - 300
Resident	<8 - <27	14 - 190
Philippines		
Migratory	6.1 - 23	12 - 94
Resident	6.3 - 32	39 - 290

From the limited studies carried out to date, it appears that seabirds that prey on organotin contaminated marine invertebrates, such as ducks, have a greater body burden of butyltins, although birds can purge butyltins by moulting and other shedding of feathers. The ecotoxicological significance of exposure of seabirds to organotins is not known although in laboratory studies, TBT has been shown to be embryotoxic, reducing hatching success and fertility and affecting enzyme and hormone activity in adult birds. Human consumption of seabirds which have invertebrate, particularly molluscan, prey sources is likely to lead to greatest exposure to dietary butyltins from seabirds.

2.3 Fish

The acute toxicity of organotins to fish has been established in laboratory studies.
A good summary of the toxicity of butyltins to marine fish is included in a paper already submitted to IMO by Japan (MEPC 41 / INF3) [28].

There is evidence of organotins having an endocrine disrupting effect in fish. TBT has been shown *in vitro* to inhibit the MFO (mixed function oxidase) system in mullet (*Mullus barbatus*) found in the western Mediterranean. This system is important in the metabolism of xenobiotics such as PAHs, PCBs

25 Guruge K S, H Iwata, H Tanaka, S Tanabe (1997) **Butyltin accumulation in the liver and kidney of seabirds** Mar Environment Res 1997b 44 (2) 191-199.

26 Senthilkumar K, S Tanabe, K Kannan and A Subramanian (1999) **Butyltin residues in migratory and resident birds collected from South India.** Toxicol and Environ Chem Vol. 68 91-104

27 Senthilkumar K, K Kannan, S Tanabe and M Prudente (1998) **Butyltin compounds in resident and migrant birds collected from Philippines.** Fresenius Environ Bull 7: 561-571 (1998)

28 Anon (1997) **Harmful Effect of the Use of Anti-Fouling Paints for Ships Call for the World-wide Ban on Every Use of Organotin-based Anti-fouling Paints for Ship Bottoms.** Submitted By Japan. MEPC 41/INF3 Appendix 5 Table 1 pp 34-36

and others leading to the detoxification or bioactivation of these compounds. The inhibition of the MFO system by TBT could have adverse effects on fish living in already polluted environments[29].

Exposure of flounder (*Platichthys flesus*) to TBT, at concentrations which were within one order of magnitude of maximum TBT levels measured in the field (experiment: 17300ng/l TBT; field: 7200ng/l TBT), caused mortality after 7-12 days, resulted in gill lesions, and induced significant reduction of the non-specific resistance. A significant decrease of the relative thymus volume was noted after exposure of flounder to TBT suggesting on endocrine disrupting effects although there were no marked effects on the specific immune system[30].

Organotins have been found in many fish species from all over the world with variable distribution in the internal organs and tissues. Organotin compounds have been found in the muscle and liver of bluefin tuna (*Thunnus thynnus*) and in the kidneys of blue shark (*Prionace glauca*) on Italy's Adriatic coast[31].

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29 Morcillo Y and C Porte (1995) **Interaction of tributyl- and triphenyltin with the microsomal monooxygenase system in molluscs and fish from the Western Mediterranean** in Proceedings of Costs v Benefits of TBT and Alternative Antifoulants - An International Conference Malta, December 1995.

30 Grinwis G C M, A Boonstra, E J Vandenbrandhof, J A M A Dormans, M Engelsma, R V Kuiper, H Vanloveren, P W Wester, M A Vaal, A D Vethaak and J G Vos (1998) **Short-term toxicity of bis(tri-n-butyltin)oxide in flounder (*Platichthys flesus*) - pathology and immune function**. Aquatic Toxicology. 1998; 42(1):15-36.

31 Kannan K, S Corsolini, S Focardi, S Tanabe and R Tatsukawa (1996) **Accumulation pattern of butyltin compounds in dolphin, tuna and shark collected from the Italian coastal waters**. Arch Environ Contam Toxicol 31, 19-23.

Table 8: Mean and range of butyltins (ng/g wet weight) in the muscle and liver of bluefin tuna and immature female blue sharks from the Mediterranean Sea[32]

Tuna:	Liver	210 (67-540)
	Muscle	62 (16-230)
Shark:	Subcutaneous fat	3.4 (1-9)
	Liver	30 (20-36)
	Kidney	140 (75-220)

Fish from the Catalan coast of the Mediterranean Sea (grey mullet *Liza aurata* and mullet *Mullus barbatus*) have been shown to bioaccumulate organotins in liver tissue, although levels were not as high as those accumulated in the same waters by mussels, snails and clams. No organotins were detected in the muscle of these fish[33].

High TBT concentrations (52.5 times those in muscle) were detected in the blood of three cultured fish species (Japanese flounder, red sea bream, yellowtails and wild flatfish. This suggests that fish serum or plasma may be a significant location of the total body burden of butyltins[34].

Butyltin concentrations were determined in tissues and stomach contents of fish collected in 41 embayments on the East, Gulf and Pacific coasts of the U.S.A. between 1986 and 1991 as part of the National Oceanic and Atmospheric Administration's (NOAA) National Benthic Surveillance Project (NBSP). A total of 108 fish liver samples from 11 fish species, and 10 composites of fish stomach contents were analysed for tetrabutylin, TBT, DBT and MBT. Many of the fish liver and stomach contents samples contained butyltins. The concentrations of butyltins in stomach contents indicated that diet is a significant route of exposure of fish to butyltins. Between 1986 and 1991 butyltin concentrations in fish generally appeared to be declining. However, no statistically significant temporal trends were observed[35].

The concentrations of butyltins in fish collected for the Gulf of Mexico during 1994 ranged between 158 and 289 ng/g wet weight showing that despite the controls on organotins put in place in 1988 in the USA, sources of TBT contamination still remain[36].

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32 Kannan K, S Corsolini, S Focardi, S Tanabe and R Tatsukawa (1996) **Accumulation pattern of butyltin compounds in dolphin, tuna and shark collected from the Italian coastal waters.** Arch Environ Contam Toxicol 31, 19-23.

33 Morcillo Y, Borghi V, Porte C (1997) **Survey of organotin compounds in the Western Mediterranean using molluscs and fish as sentinel organisms**. Arch Environment Contam. Toxicol 32, 198-203.

34 Oshima Y; Nirmala K; Go J; Yokota Y, and Koyama J (1997) **High accumulation of tributyltin in blood among the tissues of fish and applicability to environmental monitoring.** Environmental Toxicology & Chemistry. 1997; 16(7):1515-1517.

35 Krone C A, J E Stein and U Varanasi (1996) **Butyltin contamination of sediments and benthic fish from the East, Gulf and Pacific coasts of the United States.** Environmental Monitoring & Assessment. 1996; 40(1):75-89.

36 Kannan K, K Senthilkumar, B G Loganathan, S Takahaso, D K Odell and S Tanabe (1997) **Elevated accumulation of TBT and its breakdown products in bottlenose dolphins found stranded along the US Atlantic and Gulf coasts**. Environment. Sci. Tech. 31, 296-301

Fish liver samples from Australian, Papuan and Solomon Islands waters taken between 1990 and 1992, show bioaccumulation of organotins up to 570ng/g wet weight with MBT the predominant organotin species found[37].

Concentrations of butyltin residues were determined in muscle tissue of fish collected between 1990 and 1994 from local markets and sea food shops in India, Bangladesh, Thailand, Indonesia, Vietnam and Taiwan, as well as Australia, Papua New Guinea and the Solomon Islands. The results show butyltin contamination of edible fish species to be widespread.

Table 9: Concentrations of butyltins (ng/g) in fish muscle from certain Asian countries, Australia, Papua New Guinea and the Solomon Islands (ng/g)[38]

Australia

Rubberlip morwong	1.5
Shovelnose ray	20
Blue groper	47 (1.5 - 47)
Long-spined snapper	6.8
Sea mullet	1.5
Spiny tailed leatherjacket	19
Australian herring	27
Striped seaperch	25
Sea mullet	25
Black bream	26 (1.5 - 27)
Rainbow trout	21
Atlantic salmon	16 (16 - 21)
Sea mullet	12
Silver bream	12
Mud flathead	29 (12 - 29)
Silver trevally	40
Stripey	ND
Black pomfret	25
Seabass	28

Papua New Guinea

Sea mullet	9.0
Tilapia	0.13

Solomon Islands

Greenspotted kingfish	0.2
Indian mackerel	1.4
Paddletail snapper	0.79 (0.2 - 1.4)

37 Kannan K, S Tanabe, R Tatsukawa and R J Williams (1995) **Butyltin residues in fish from Australia, Papua New Guinea and the Solomon Islands** Intern J Environ Anal Chem Vol 61 pp 263-273

38 Kannan K, S Tanabe, H Iwata and R Tatsukawa (1995) **Butyltins in muscle and liver of fish collected from certain Asian and Oceanian countries**. Environmental Pollution. 1995; 90(3):279-290

India

Scombrid	0.69
Catfish	11
Jawfish	9.2
Sciaenid fish	ND
Indian mackerel	1.7 (ND - 11)
Sea mullet	18
Pearl spot	ND
Catla	7.0
Indian mackerel	56
Silver pomfret	79 (7.0 - 79)
Catla	41
Black bream	33
Threadfins	47
Perch	40 (33 - 47)

Bangladesh

Flounder	92
Catla	190
Hilsa	15
Aor	0.47 (0.47 - 190)

Vietnam

Silver carp	ND
Carp	ND
Bream	0.93
Sea mullet	ND
Perch	0.24
Tilapia	0.23
Striped snake-head fish	ND

Indonesia

Big-eyed scad	19
Deep-bodied crucian carp	0.41 (0.41 - 19)

Taiwan

Tilapia	0.49
Milkfish	0.96
Seabream	18

Butyltin compounds were detected in most of the samples which suggested widespread contamination in Asia and Oceania. Corresponding liver samples of fish collected in Australia, Papua New Guinea and the Solomon Islands were analysed to obtain information on partitioning of butyltin compounds between muscle and liver tissues. The concentrations of butyltin compounds were, on average, an order of magnitude higher in liver than in muscle. Butyltin concentrations in fish from Asia and Oceania were lower than those reported for Japan, Canada and the USA[39].

Organotin contamination in deep-sea ecosystems has been detected in Suruga Bay, Japan. Organisms collected between 135 and 980 m in the aphotic bathyal zone were compared with those collected from shallow waters. Total butyltin concentrations in the tissues of deep-sea fish, crustaceans, cephalopods, echinoderms, and gastropods were up to 980, 460, 460, 130, and 21 ng/g wet weight, respectively. These levels were lower than those in shallow-water organisms from the same bay but comparable to those reported in industrialised areas like Tokyo Bay, suggesting the expansion of butyltin pollution to deep-sea ecosystems. TBT was the predominant organotin, rather than DBT or MBT, except in cephalopods, suggesting a fresh input of TBT into the deep-sea environment[40].

In 1990, 58 fish from nine species from Gdansk Bay, Baltic Coast of Poland were sampled and tested for butyltin residues.

Table 10: Concentrations of butyltins in fish from the Southern Baltic Sea.

	Total butyltins (range in brackets) (ng/g wet weight)
Flounder	316
Herring	40
Eel	188
Sea Trout	51 (45-57)
Turbot	39
Cod	19 (14-24)
Eelpout	130
Pikeperch	455
Mackerel	27 (23-20)

The extent of contamination of these species has raised questions of the level of exposure of the fish-eating Polish human population[41].

Repeat studies of Gdansk Bay fish sampled during 1997 showed that total butyltin concentrations in fish tissues remain high in the Southern Baltic Sea[42]. Higher levels than both Mediterranean and Oceanian water were found in the Southern Baltic Sea. Levels of butyltin ranged up to 4600ng/g wet

39 Kannan K, S Tanabe, H Iwata and R Tatsukawa (1995) **Butyltins in muscle and liver of fish collected from certain Asian and Oceanian countries**. Environmental Pollution. 1995; 90(3):279-290

40 Takahashi S, S Tanabe and T Kubodera (1997) **Butyltin residues in deep sea organisms collected from Surnga Bay, Japan.** Environmental Science & Technology. 1997; 31(11):3103-3109

41 Kannan K and J Falandysz (1997) **Butyltin residues in sediment, fish, fish-eating birds, harbour porpoise and human tissues for Polish Coast of the Baltic Sea**. Mar Poll Bull vol. 34 no 3 pp 203-207

42 Senthilkumar K, C A Duda, D L Villeneuve, K Kannan, J Falandysz and J P Giesy (1998) **Butyltin compounds in sediment and fish from the Southern Baltic Sea, Poland, 1997-1998**. Environmental Science and Pollution Research (accepted for publication)

weight in herring liver[43] and up to 102ng/g wet weight in eel-pout from the German North Sea[44].

Table 11: Concentration (ng/g wet weight) of butyltin compounds in selected tissues of fish collected from the Southern Baltic Sea and Vistula River[45]

Species	Tissue	Total butyltins
Herring	Egg	370
	Liver	4800
	Muscle	78
Ruff	Egg	170
	Liver	1200
	Muscle	44
Smelt	Egg	16
	Liver	440
	Muscle	170
Flounder	Muscle	83
Turbot	Muscle	110
Brown Trout	Muscle	78
Roach	Muscle	3300
Burbot	Egg	39
Burbot	Liver	32
Perch	Liver	410
Roach	Muscle	100

In conclusion, it is clear that organotin contamination of edible fish species is widespread across the world. The toxicity of organotins to fish has been established. There is also evidence of organotins having an endocrine disrupting effect on fish. The widespread contamination of fish shows that concern that organotins may threaten humans consuming large quantities of fish in their normal diet is legitimate; particular risk may be incurred in populations consuming fish liver as organotins appear to be concentrated in the liver at an order of magnitude greater than muscle.

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43 Senthilkumar K, C A Duda, D L Villeneuve, K Kannan, J Falandysz and J P Giesy (1998) **Butyltin compounds in sediment and fish from the Southern Baltic Sea, Poland 1997-98** Environmental Science and Pollution Research IN PRESS

44 Shawky S and H Emons (1998) **Distribution pattern of organotin compounds at different trophic levels of aquatic organisms.** Chemosphere 36 (3) 523-535

45 Senthilkumar K, C A Duda, D L Villeneuve, K Kannan, J Falandysz and J P Giesy (1998) **Butyltin compounds in sediment and fish from the Southern Baltic Sea, Poland, 1997-1998.** Environmental Science and Pollution Research (accepted for publication)

3 Intake of organotins by Humans

It is known that organotins have endocrine disrupting ability. For example, researchers from Murray State University, USA have shown that butyltins disrupt the critical function of human immune cells, particularly killer cells. Indeed concerning exposure of humans to butyltins is shown by the finding of biologically significant levels of butyltins in random human blood samples[46].

A tolerable daily intake (TDI) of 15ug TBT per person per day for a 60kg person has been set out on the basis of reduction of immune function[47]. There is no recognised TDI for total butyltins or other organotins. Nevertheless, there have been various attempts to analyse whether the level of organotin intake by humans is at a level to cause concern. Most have suggested that organotin contamination as a result of eating contaminated marine species falls short of reaching tolerable or acceptable daily intakes. However other studies suggest that TDIs hare being reached in parts of the world and by particular communities consuming large quantities of seafood.

Concentrations of butyltin residues have been determined in muscle tissue of fish collected from local markets and sea food shops in India, Bangladesh, Thailand, Indonesia, Vietnam, Taiwan, Australia, Papua New Guinea and the Solomon Islands. Butyltin concentrations in fish from Asia and Oceania were lower than those reported for Japan, Canada and the USA. Although the number of samples analysed from each country was small, it has been tentatively suggested by these studies that intake of butyltins by humans via consumption of fish in these countries was < 25% of the tolerable daily intake of 250 ng / kg body weight / day [48].

Concentrations of butyltins determined in the muscle and liver of those fish collected from Australia, Papua New Guinea and the Solomon Islands between 1990 and 1992 were examined, as part of this study. The daily dietary intake of butyltins by Australians via these fish was estimated to be 337-416 ng / person / day, lower than is believed to cause health problems[49].

46 Anon (1999) **Chemicals in boat paint, household items blocks cancer fighting cells.** Anaheim, California March 24th 1999 Environmental News Service.

47 Penninks A H (1993) **The evaluation of data-derived safety factors for bis(tri-n-butyltin)oxide.** Food Addit. Contam 10, 351-361.

48 Kannan K, S Tanabe, H Iwata and R Tatsukawa (1995) **Butyltins in muscle and liver of fish collected from certain Asian and Oceanian countries.** Environmental Pollution. 1995; 90(3):279-290

49 Kannan K, S Tanabe, R Tatsukawa and R J Williams (1995) **Butyltin residues in fish from Australia, Papua New Guinea and the Solomon Islands.** International Journal of Environmental Analytical Chemistry. 1995; 61:263-273.

Table 12: Estimated average daily intake of butyltin compounds by Australians at different age groups (ng per person per day) via fish muscle consumption (MBT, DBT and TBT levels are shown in brackets, respectively)[50]

Age Group (years)	Fish consumption (g/day)	Butyltin intake (ng/person/day)
15	17	337 (265, 24, 48)
25-34	18	356 (281, 25, 50)
Average	21	416 (328, 29, 59)

In 1993, the Australian National Food Authority concluded that there was no apparent health risk to the public from the normal or excessive consumption of seafood contaminated with TBT[51].

Daily butyltin and triphenyltin (TPT) intakes from the Shiga Prefecture of Japan have also been investigated. Foods tested included rice, cereals, potatoes, sugar, cakes, fats and oils, beans, fruits, vegetables, seaweeds, seasonings, fish and shellfish, meat, eggs, milk and dairy produce. Daily intakes of TBT and TPT detected by the duplicate portion method were found to be 4.7 and 0.7 ug/g in 1991 and 2.2 and 0.7 ug/g in 1992. Corresponding figures for the market basket methods were 6.9, 5.4, 6.7 and 1.3 ug/g respectively. All values were less than the 80ug/50kg bodyweight Japanese Welfare Ministry acceptable daily intake for TBT and the 40ug/50kg bodyweight FAO/WHO level for TPT, although both these levels had been exceeded during 1988-1991[52].

It has been concluded in papers submitted to IMO by CEFIC that health effect are unlikely from exposure to TBT in market-bought seafood caught during 1989-1990 in the USA. The submission examined average per capita sea food consumption rates for a number of countries, based on seafood purchased in 1997 in Stockholm (Sweden), London (England), Marseille (France), Singapore, Ulsan (Korea), Sydney (Australia), Galveston (USA) and Halifax (Canada). The amount of TBT ingested was considered unlikely to exceed proposed threshold for chronic effects, suggesting negligible risk to the average consumer. However this study does not consider rates of fish consumption by artisanal or subsistence fishers which would be higher than the estimates used in this study[53].

However some recent work does appear to suggest that the consumption of contaminated fish is posing a real threat to humans. The results of analysis of fish from Gdansk Bay sampled in 1990 raised questions as to the safety of fish consumption by the Polish human population. Herring cod and flounder constitute 90% of the Baltic fishery consumed by Poles. Based on the 50g/day consumption of fish by Poles, the estimated daily intake of butyltins by Poles is 770-22800 ng/person. The upper

50 Kannan K, S Tanabe, R Tatsukawa and R J Williams (1995) **Butyltin residues in fish from Australia, Papua New Guinea and the Solomon Islands.** International Journal of Environmental Analytical Chemistry. 1995; 61:263-273.

51 Australian and New Zealand Environment and Conservation Council (1995) **Impacts on the Marine Environment from Shipping: Attachment 4 Report on Antifouling Paint - Organotins** http://www.environment.gov.au/portfolio/epg/pubs/map_att4.html 29th November 1996

52 Tsuda T, T Inoue, M Kojima and S Aoki (1995) **Daily intakes of TBT and TPT compounds from meals.** Journal of AOAC International 1995 78(4) 941-943

53 CEFIC (1997) **Harmful effects of the Use of Antifouling Paints for Ships - An update about toxicology and ecotoxicology of TBT** Submitted to IMO by CEFIC MEPC41/INF6 31st December 1997

value exceeds the tolerable daily intake (TDI) for an individual weighing 60kg of 15000ng/day[54]. Butyltin concentrations in human liver from the Gdansk Bay area from 1994 ranged from 2.4 to 11ng/g wet weight[55].

There has been some debate over the methodology used in this study, and questions over the validity of, for example, using a Tolerable Daily Intake derived for TBT to cover all butyltin residues which, of course, have differential toxicity[56]. Nevertheless, it appears that when the consumption by Poles of fish liver, which can contain up to ten times the concentration of butyltins as fish muscle, and the high intake of fish in the diet in the Gdansk Bay area (250 - 350 g /day) are taken into account, there are indeed serious grounds for concern[57].

Repeat studies of Gdansk Bay fish sampled during 1997 showed that total butyltin concentrations in fish tissues remain high in the Southern Baltic Sea. The estimated daily intake of butyltins in the fish-eating Polish human population ranged from 2.2 to 164 ug/person with the intake for roach exceeding the TDI of 15ug TBT per 60kg person per day. Furthermore due to the preferential accumulation of butyltins in fish liver and the consumption of fish liver (cod, burbot and salmon) by Poles, as well as reports of high fish consumption (up to 250g fish consumed per person per day near the city of Gdansk), the intake of TBT may be greater than this figure. At 250g/day fish consumption levels, the TDI of 15ug was exceeded for all the regularly consumed marine Gdansk Bay fish species studied[58].

It is also evident that caution should be exercised when limiting considerations of total dietary intake of organotins to that from only marine food sources. As well as for their antifouling properties, organotin compounds are used in a variety of consumer and industrial products including agricultural pesticides, wood preservatives, and as stabilisers in PVC plastic. Octyltin, butyltin, and phenyltin derivatives are permitted for use as stabilisers in plastic products which come into contact with food. TBT is also applied as disinfectants in waxes, polishes, sprays and in laundry washes may cause contamination of sewage effluents and sludge. Industrial discharges of TBT, used as a slimicide in the paper industry and for textile and lumber treatment and in cooling water treatment, are further sources of sewage sludge contamination. Since sludge is used as a fertiliser in agriculture in many parts of the world, butyltin species could be transferred to soils. Recent studies have shown the presence of butyltins in a variety of household textiles including nappy covers, sanitary panties, socks, etc.

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54 Penninks A H (1993) **The evaluation of data-derived safety factors for bis(tri-n-butyltin)oxide.** Food Addit. Contam 10, 351-361

55 Kannan K and J Falandysz (1997) **Butyltin residues in sediment, fish, fish-eating birds, harbour porpoise and human tissues for Polish Coast of the Baltic Sea**. Mar Poll Bull vol. 34 no 3 pp 203-207

56 Robinson S, J Volosin, J Keithly and R Cardwell (1999) **Comment on Butyltin residues in sediment, fish, fish-eating birds, harbour porpoises and human tissue from the Polish coast of the Baltic Sea** Mar Poll Bull Vol 38 No 1 pp 57-61

57 Kannan K and J Falandysz (1999) **Response to Comment on Butyltin residues in sediment, fish, fish-eating birds, harbour porpoises and human tissue from the Polish coast of the Baltic Sea** Mar Poll Bull Vol 38 No 1 pp61-63

58 Senthilkumar K, C A Duda, D L Villeneuve, K Kannan, J Falandysz and J P Giesy (1998) **Butyltin compounds in sediment and fish from the Southern Baltic Sea, Poland, 1997-1998.** Environmental Science and Pollution Research (accepted for publication)

Although the contamination of marine food by butyltins may be more widespread, the contamination of food other than marine food cannot and should not be ruled out [59].

4 Discussion and Conclusions

4.1 The ecotoxicological impact of organotins on fish, sea birds and marine mammals is not as well studied as the impact on marine invertebrates, particularly gastropod molluscs. Nevertheless, organotins have been shown to be toxic to fish, birds and mammals and have also been demonstrated to have endocrine disrupting properties in these animals.

4.2 Organotins are also bioaccumulative in certain marine species, some of which are food species used by humans. Organotins have been detected in a range of edible marine species, including fish, birds and mammals as well as invertebrates such as molluscs, crustaceans and cephalopods. There is justifiable concern over the level of exposure of humans to organotins consumed in edible marine species.

4.3 Marine mammals belonging to a range of species have been found to be contaminated with organotins. Butyltin compounds including TBT, DBT and MBT have been detected in almost all marine mammal liver samples from wherever in the world they are taken. This illustrates the worldwide distribution of organotins in the oceans. The elevated residues detected in coastal species and low concentrations found in off-shore species indicate a high degree of organotin contamination in coastal waters. Mammals inhabiting waters of developed nations are, in general, found to contain higher concentrations compared with those collected from the waters of developing countries. Some mammals such as the sea lion have the ability to degrade or expel butyltins for the body, while others, such as dolphins exhibit age-dependent biomagnification of butyltins. High doses of organotins have been shown to damage the central nervous system and reproductive mechanisms in mammals. TBT is an endocrine disrupting chemical in mammals. It is likely that negative effects are occurring to marine mammals, including sea otters, from exposure to organotins.

It is also likely that organotin contamination is being passed to humans consuming marine mammals as a significant part of their diet. Although not all marine mammals are utilised for human food, a number of different seals, whales and to a lesser extent dolphins are consumed by humans, particularly, although not exclusively, in artisanal and subsistence fishing communities. In some populations, marine mammals provide a very significant proportion of dietary protein and fat intake.

4.4 There is less data on organotin residues and their impact in seabirds than for marine mammals. From the limited studies carried out to date, it appears that seabirds, such as sea ducks, that prey

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59 Kannan K, S Tanabe and R Tatsukawa (1995) **Occurrence of butyltin residues in certain foodstuffs.** Bulletin of Environmental Contamination & Toxicology. 1995; 55(4):510-516

on organotin contaminated marine invertebrates have a greater body burden of butyltins, although birds can purge butyltins by moulting and other shedding of feathers. Nevertheless, high levels of organotins have been found in marine birds in coastal locations. The ecotoxicological significance of exposure of seabirds to organotins is not known although in laboratory studies, TBT has been shown to be embryotoxic, reducing hatching success and fertility and affecting enzyme and hormone activity in adult birds. Human consumption of seabirds which have invertebrate, particularly molluscan, prey sources is likely to lead to greatest exposure to dietary organotins from seabirds.

4.5 Organotins have been found in many fish species from all over the world with variable distribution in the internal organs and tissues. The toxicity of organotins to fish has been established. There is also evidence of organotins having an endocrine disrupting effect on fish. A good summary of the toxicity of organotins to marine fish is included in a paper already submitted to IMO by Japan (MEPC 41 INF3).

The widespread contamination of many fish shows that concern that organotins may threaten humans consuming large quantities of fish in their normal diet is legitimate. Particular risk may be incurred in populations consuming fish liver as butyltins appear to be concentrated in the liver at an order of magnitude greater than muscle.

4.6 Organotins have endocrine disrupting ability in humans. Butyltins disrupt the critical function of human immune cells, particularly killer cells. Recent findings have revealed biologically significant levels of butyltins in random human blood samples.

A tolerable daily intake of 15ug TBT per person per day for a 60kg person has been set out on the basis of reduction of immune function. There is no TDI for total butyltins or all organotins. Nevertheless, there have been various attempts to analyse whether the level of organotin intake by humans is at a level to cause concern. Most have suggested that levels as a result of eating contaminated marine species falls short of reaching tolerable or acceptable daily intakes. However recent studies suggest that TDIs have being reached in parts of the world and by particular communities consuming large quantities of seafood. Exposure to other sources of organotins in addition to dietary intake from marine species cannot be ruled out.

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