Chapter 6
Organotin Contamination in Deep Sea Environments

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Abbreviations BMF: Bio magnification factor; CHL: Chlordane compounds; DBT: Dibutyltin; DDTs: DDT and its metabolites; DPT: Diphenyltin; HCH: Hexachlorobenzene; MBT: Monobutyltin; MPT: Monophenyltin; OT: Organotin; PAH: Polycyclic aromatic hydrocarbon; PCB: Polychlorinated biphenyl; POPs: Persistent organic pollutants; ROVs: Remotely operated vehicles; SPM: Suspended particulate material; TBT: Tributyltin; TPT: Triphenyltin

6.1 Introduction

Deep sea environments are divided into the bathyal zone (200–2,000 m), the abyssal (2,000–6,000 m) and the hadal zone (over 6,000 m). These zones cover the largest part of the ocean biome (more than 80%). Until now, it was considered that these zones were deserts because sunlight could not reach to such depths and pressures were too high for biota. Advances in deep sea submersibles and image capturing technologies are now increasing the opportunities for marine biologists to observe

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and uncover the mysteries of the deep ocean realm. Remotely operated vehicles (ROVs) have been used underwater since the 1950s. ROVs are basically unmanned submarine robots with umbilical cables used to transmit data between the vehicle and researcher for remote operation in areas where diving is constrained by physical hazards. ROVs are often fitted with video, cameras, mechanical tools for specimen retrieval and measurements. Subsequently, manned deep sea submersibles have been developed and research has progressed. Although the deep sea is in total darkness, is extremely cold, and subjection to great pressure, the marked development of bathyscaphes has revealed the presence of many deep-sea organisms such as bivalves and gastropods in water depths of 3,000 m and more, and has permitted the collection of sediment and marine organisms (e.g. Endo et al. 1999; Okutani et al. 2002; Okutani and Iwasaki 2003).

The contamination of deep-sea ecosystems by man-made chemicals has also been clarified by progress in diving technology. Organochlorine insecticide residues were measured in the livers of *Antimore rostrata*, a deep sea fish collected from 2,500 m in 1972, 1973, and 1974 off the east coast of the United States (Berber and Warlen 1979). Subsequently, metals, polycyclic aromatic hydrocarbons (PAHs), polychlorinated biphenyls (PCBs) and chlorinated pesticides in were determined in tilefish (*Lopholatilus chamaeleonticeps*) collected from Lydonia Canyon (on the Georges Bank) in 1981–1982 (Steimle et al. 1990). Persistent organochlorines such as PCBs, DDT and its metabolites (DDTs), chlordane compounds (CHLs), and hexachlorobenzene (HCH) were detected in deep-sea organisms from a water depth of 180–980 m in Suruga Bay (Lee et al. 1997), whilst PCBs have also been detected (22 mg kg^{-1}) along with DDTs (13 mg kg^{-1}) in amphipods collected from a water depth of 2,075 m in the Arctic Ocean (Hargrave et al. 1992). More recently, detection of persistent organic pollutants has been reported in many kind of samples from various water depths (e.g., Takahashi et al. 1997a). It has thus been concluded that persistent organic pollutants (POPs) and various other contaminants can be transferred to deep-sea areas where they may be accumulated by deep-sea organisms.

It is well known that organotins (OTs) cause advertise effects in marine organisms (Gibbs and Bryan 1986; Gibbs et al. 1991; Ohji et al. 2002) and may persisted in sediments for a long time (Dowson et al. 1993; Harino et al. 1999; see also Chapter 6, this volume). Therefore the current status of OTs and their effects in deep-sea organisms are of much concern. In this chapter, recent findings on the contamination of deep-sea environments by OTs are summarized. Throughout, concentrations of OTs are expressed as Sn^{4+}.

6.2 Details of Deep-Sea Areas

There are few papers on the contamination of OTs in deep sea environments, because of the difficulty and costs in taking samples in the deep sea To our knowledge, only six such surveys (of OTs) have been performed in the Mediterranean Sea

(Michel and Averty 1999; Borghi and Porte 2002), and at five sites around Japan – Yamato Bank (Ikeda et al. 2002), Tosa Bay (Takahashi et al. 2001), Suruga Bay (Takahashi et al. 1997b), off-Tohoku (De Brito et al. 2002) and the Nankai Trough (Harino et al. 2005). The Mediterranean Sea bordered many countries each housing considerable numbers of ports, harbours and marinas. The latter have increased substantially in recent years, especially, in the northwestern Mediterranean Sea. There is also considerable commercial and naval shipping traffic.

The Yamato Bank is located in the central part of the Sea of Japan between China and Japan, and has become an eminent fishing ground. The central part of the Yamato Bank is divided by a ravine of 2,000 m. Suruga Bay is located near the central part of the Pacific coast of Honshu and has a deep and narrow trough along its long axis. The bathyal zone (200–1,000 m) of the eastern continental slope of the bay has a steep but simple physiography. There are many open ocean fisheries bases and some paper mill industries in the inner part of the bay. Tosa Bay is located along the Pacific coast of Kochi Prefecture and has an entrance which is deep and opens into the Pacific Ocean. The central part of the continental slope of the bay is at a depth of 25–800 m. The hydrological circulation within the bay is strongly influenced by the inflow of the oceanic water, the Kuroshio current, resulting in effective water exchange in and out of the bay. Human influences are not intensive along the coast of Tosa Bay, except for the harbor of Kochi Port which houses a significant number of maritime and industrial activities. Off-Tohoku is located in the northwestern Pacific and human activities are relatively low along the adjacent coastal area of northern Japan: here there are major ocean currents, Oyashio (northward cold water) and Kuroshio (southward warm water); which together contribute to, Kuroshio – Oyashio interfrontal zone along the Pacific coast of northern Japan. The continental slope in this region is at a depth of 150–1,300 m. The Nankai Trough, referring to the western edge of the smaller Philippine Sea Plate which is in contact with Japan, is located 100 km off the southern coast of Japan and has a water depth of 3,000–4,000 m. Fishing is the main industry of Cape of Muroto, the closest point to Nankai Trough, and whilst many fishing boats from other countries also frequent these waters.

6.3 The Concentrations of Organotins in Deep Sea Environments

The concentrations of OTs at these deep sea sites are summarized in Table 6.1. Three vertical profiles between 25 and 2,500 m in water columns from the northwestern Mediterranean Sea were surveyed by Michel and Averty (1999). The vertical profile of salinity and tributyltin (TBT) concentrations at the three stations are shown in Fig. 6.1. Highest levels occurred in high salinity surface water. The inflow of sea water at a depth of around 500 m is confirmed by the vertical profile of salinity and the lowest TBT concentrations occurred at this level. Below 500 m, a secondary peak in TBT concentrations was observed at 1,200 m, and thereafter,

Table 6.1 The concentrations of OTs in deep sea environment

Location	Water depth	Sample	MBT	DBT	TBT	MPT	DPT	TPT
Northwestern Mediterranean[a]	500–2,500	Water	–	–	<06–16	–	–	–
Northwestern Mediterranean[b]	1,008–1,816	Fish (liver)	<1.0–72	6.1–82	2–64	<1–190	<1–110	<1.4–240
		Fish (muscle)	<1.0	<1–9.2	<1	<1	<1	<1–4.3
		Fish (gill)	<1.0–1.9	<1–6.8	<1–18	<1	<1	<1.4–71
		Fish (disgestive grand)	<1.0–2.2	<1–3.5	<1–20	<1	<1–18	<1.4–49
Yamato Bank, Japan[c]	350–400	Water	–	–	246–328	–	–	<0.3
	350–401	Sediment	–	–	2.3–6.6	–	–	1.3–2.3
	200–740	Fish	–	–	1.5–34	–	–	14–75
	350–980	Crustaceans	–	–	0.74–32	–	–	44–156
	200–540	Cephalopods	–	–	28–43	–	–	27–48
Suruga Bay, Japan[d]	200–740	Fish	<10–160	<2.0–350	<0.8–280	–	–	–
	350–980	Crustaceans	<10–17	<2.0–47	3.2–150	–	–	–
	200–540	Cephalopods	<10–61	<2.0–82	1.8–98	–	–	–
	135–350	Echinoderms	<2.7–14	2.8–5.6	2.3–53	–	–	–
	135–350	Gastropods	<10	4.2	5.3	–	–	–
North Pacific, off-Tohoku, Japan[e]	250–1,000	Fish	<1.2–28	<0.2–27	0.1–180	–	–	–
	150–1,000	Cephalopods	1.0–7.5	0.5–4.4	0.9–13	–	–	–
	250–2,000	Crustaceans	<1.2–18	<0.2–1.4	<0.1–5.3	–	–	–
	250–400	Gastropods	<1.2–5.4	<0.2–4.6	0.6–1.9	–	–	–
	2,000	Sea star	<1.2	<0.2	0.8	–	–	–

Water; ng l^{-1}, sediment; μg kg^{-1} dry, biological sample; μg kg^{-1} wet

[a]Michel and Averty (1999)

[b]Borghi and Porte (2002)

[c]Ikeda et al. (2002)

[d]Takahashi et al. (1997b)

[e]De Brito et al. (2002)

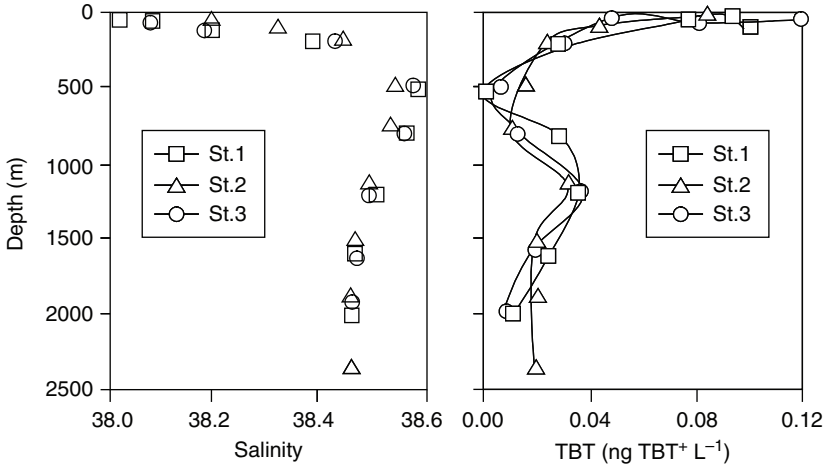

Fig. 6.1 The vertical profile of TBT and salinity in Mediterranean Sea (Borghi and Porte 2002)

contamination declined progressively down to 2,500 m. Concentrations of TBT in deep sea fish tissues (liver, muscle, gill and digestive grand) from the northwestern Mediterranean Sea were in the range <1–64 μg kg^{-1} (Borghi and Porte 2002). Despite the lower concentrations of TBT in these deep sea fish, in comparison with shallow water fish collected in harbors and coastal areas, these results highlight the transport of BT contamination to deep-sea organisms. Furthermore, the TBT residues recorded in deep-sea organisms were within the estimated effective concentrations (8–40 μg kg^{-1}) based on chronic toxicity endpoints in molluscs and were close to concentrations which exert cytotoxic effects in fish hepatoma cells. Interestingly, triphenyltin (TPT) concentrations in *A. rostratus* and *M. moro* from deep waters were much higher than those reported in coastal fish from the area.

The locations of the five areas near Japan in which surveys of OT contamination in the deep sea were conducted are shown in Fig. 6.2. Kono et al. (2004) measured TBT and TPT in water from the Yamato Bank at a depth of 350–400 m. The concentrations of TBT were in the range 0.2–0.3 ng l^{-1}, which were lower than in coastal waters; TPT was not detected in this area (<0.3 ng l^{-1}). TBT in sediment was detected in the range 2.3–6.6 μg kg^{-1} dry weight, again lower than those in coastal area however, TPT concentrations in deep sea sediments ranged between 1.3 and 2.3 μg kg^{-1} dry weight, comparable to values in coastal areas. TBT and TPT concentrations in sediment were 10^5 times higher than those in seawater.

OT concentrations have been measured in marine organisms from Suruga Bay, off-Tohoku and Tosa Bay. In the bathyal region of Suruga Bay, maximum TBT concentrations in the tissues of deep sea fish, crustaceans, cephalopods, echinoderms, and gastropods were 280, 150, 98, 53 and 5.3 μg kg^{-1}, respectively. Off Tohoku, the maximum concentrations of TBT were 180, 13, 5.3, 1.9 and 0.8 μg kg^{-1} in fish, cephalopods, crustaceans, gastropods and sea star, respectively. Maximum concentrations of TBT in fish, crustaceans, cephalopods and echinoderms from Tosa Bay

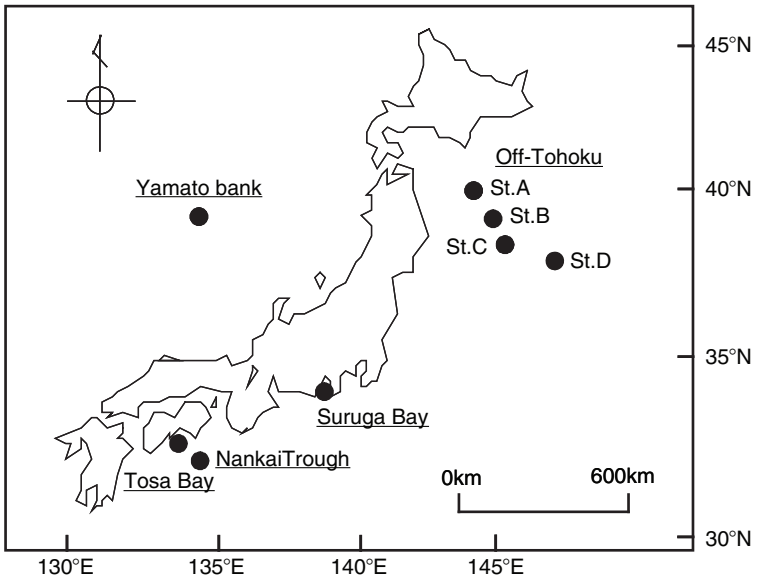

Fig. 6.2 Deep sea sampling sites for organotins, Japan

were 16, 4.5, 2.9 and <0.8 μg kg^{-1}, respectively. In these deeper bays, TBT concentrations in aquatic organisms were apparently lower than those in various coastal waters. However, their presence in tissues indicates the influence of industrial and human activities and that OT contamination is exported to the deep sea environment. Differences in BTs concentrations in myctophid fish, which undergo species-specific diel vertical migration between mesopelagic (200–1,000 m) and epipelagic (<100 m) zones, have been described by Takahashi et al. (2000). Myctophid fishes can be grouped into vertical migratory species, semi - migratory species, and non-migratory species. BTs concentrations in migratory and semi-migratory species were higher than those in non-migratory species, which may be due to higher exposure of BTs in surface water.

In the Naikai Trough, the deepest region sampled, gastropods (*Colliloconcha nankaiensis*), sea cucumbers (*Psychropotes verrucosa*), galatheid crabs (*Munidopsis albatrossae* and *Munidopsis subsquamosa*), bivalves (*Clyptogena tsubasa* and *Clyptogena nautilei*) were collected, along with sediments (from a depth of up to 1 cm and a depth of up to 15 cm below the sediment surface). BT concentrations in the 0–1 cm and 0–15 cm sediment cores were 41 and 21 μg kg^{-1} dry, respectively. The sedimentation rate near the sampling point is 1.09 mm $year^{-1}$ (Iwai et al. 2004). This result therefore indicates a recent increase in BT input. The compositions of BTs in the 0–1 cm surface samples were similar to those in sub-surface samples from a depth of up to 15 cm in the sediment core (Fig. 6.3). The PT concentration in the 0–1 cm surface sediment (0.028 mg kg^{-1} dry) was lower than at 0–15 cm depths (0.052 mg kg^{-1} dry). Moreover, the proportion of DPT and MPT, which are degradation products of TPT, was higher in surface layers (Fig. 6.3). This profile

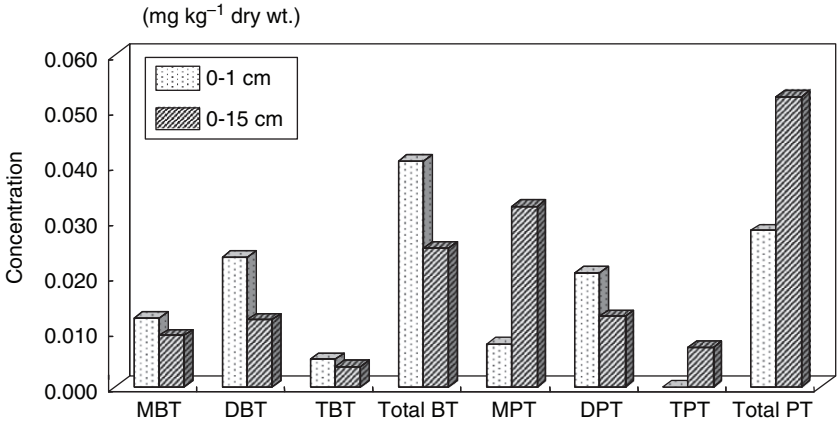

Fig. 6.3 The composition of BTs and PTs in sediment from Nankai Trough

implies a recent reduction of PT concentrations in deep-sea areas, consistent with observations in coastal areas. TBT and TPT in sediment from Nankai Trough were detected in the range 4–5 μg kg^{-1} dry weight and <1–7 μg kg^{-1} dry weight, respectively. It was reported by Ministry of the Environment, Japan (2003) that the geometric means of TBT and TPT concentrations in coastal areas of Japan were 9.4 and 1.2 μg kg^{-1} dry, respectively. The levels of TBT and TPT in sediment from deep-sea areas were thus similar to those in coastal areas, confirming the spread of OT contamination from coastal areas.

OT concentrations measured in the six species of deep-sea organism from Nankai Trough are shown in Table 6.2. Means of BT concentrations in *C. nankaiensis*, *P. verrucosa*, *M. albatrossae*, *M. subsquamosa*, *Cl. tsubasa*, and *Cl. nautilei* were, respectively, 0.089, 0.057, 0.018, 0.016, 0.019 and 0.026 mg kg^{-1} wet weight. The corresponding PT concentrations in those animals were 0.212, 0.363, 0.166, 0.186, 0.030 and 0.025 mg kg^{-1} wet weight, respectively. High concentrations were thus observed in gastropods and sea cucumbers.

6.4 Bio-Concentrations of Organotin Compounds in Deep Sea Organisms

Borghi and Porte (2002) estimated the food chain magnification of OTs in *M. moro* and *L. lepidon*. The biomagnification factor (BCF) is obtained by dividing the concentration in the whole body by the concentration in contents of their digestive tube. BCF for TBT ranged from 0.02 in *M. moro* to 0.04 in *L. lepidion*, whereas TPT ranged from 0.27 in *M. moro* to 0.93 *L. lepidion*. They clearly indicate the biodegradable nature of TBT in comparison with TPT, though on this basis neither appear to be biomagnified in the food chain.

Table 6.2 The concentrations of OTs in deep sea environment

Location	Water depth	Sample	MBT	DBT	TBT	MPT	DPT	TPT
Tosa Bay, Japan[a]	150–400	Fish	<1.4–8.2	<0.8–4.2	0.9–16	–	–	–
	150–400	Crustaceans	<1.4–2.4	<0.8–1.0	<0.8–4.5	–	–	–
	100–400	Cephalopods	<1.4–7.5	<0.8–1.8	2.0–2.9	–	–	–
	100–800	Echinoderms	<1.4–2.2	<0.8–0.8	<0.8	–	–	–
Nankai Trough, Japan[b]	3,553–3,571	Sediment	9–12	12–23	4.0–5.0	8–33	13–21	<1.0–7.0
	3,536–3,587	Bivalves	1–52	2–35	7.0–89	13–48	<1.0	<1.0
	3,540–3,575	Gastropods	10–110	5–56	2.0–34	73–330	<1.0–2.0	<1.0–13
	3,571–3,574	Galatheid	10	6	3.0	140	<1.0	26
	3,571–3,575	Crab	10	6	1.0–3.0	140–180	<1–1	6.0–26
	3,655	Sea cucumber	44	9	3.0	360	5.0	3.0

Water; ng l^{-1}, sediment; μg kg^{-1} dry, biological sample; μg kg^{-1} wet

[a]Takahashi et al. (2001)

[b]Harino et al. (2005)

Kono et al. (2004) reported the concentration of OTs in relation to trophic level in deep sea organisms from Yamato Bank (Fig. 6.4). Trophic level was classed by stomach contents of the predators: Trophic level 1 was composed of organisms ingesting mainly detritus organisms; Trophic level 2 was composed of organisms ingesting polychaeta,

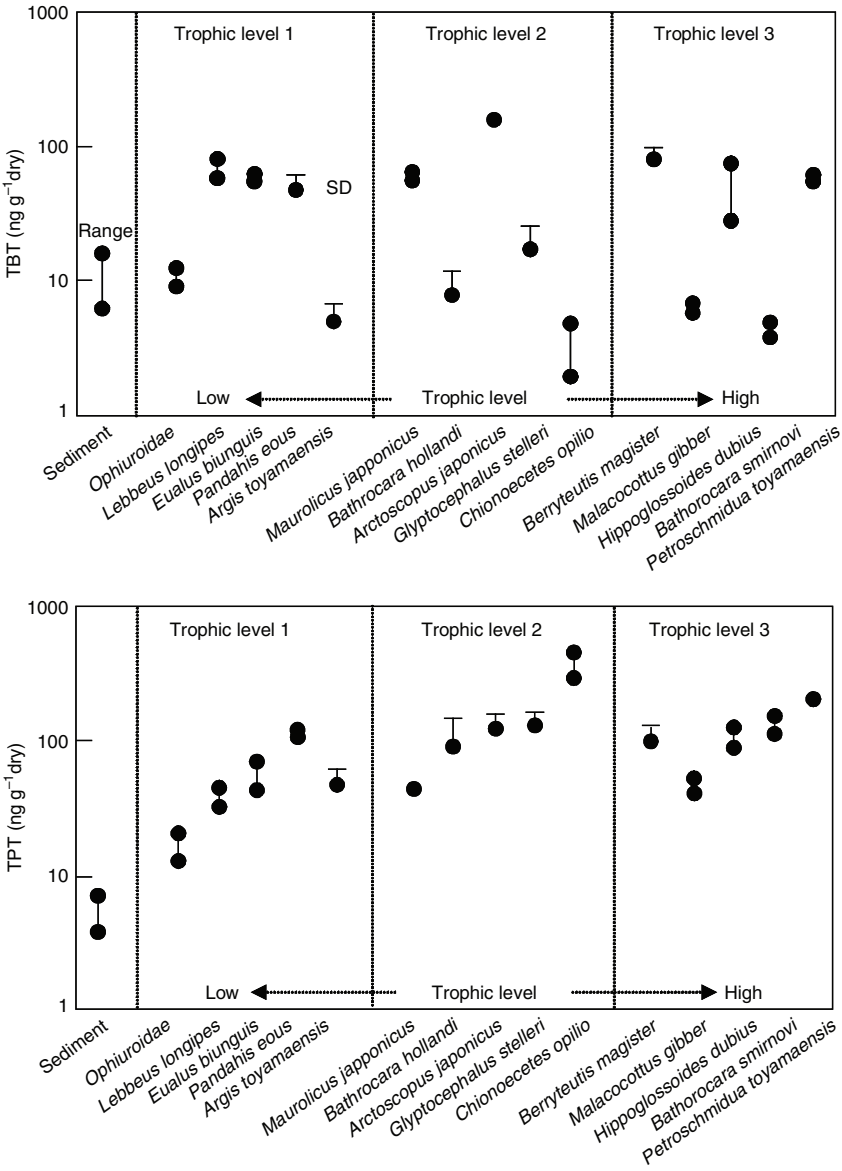

Fig. 6.4 The relationship between TBT or TPT concentrations and trophic level in Yamato Bank (Ikeda et al. 2002)

amphipoda, euphausia, and ophiuroidae; whilst Trophic level 3 was composed of organisms ingesting fish, decapods and squid. According to this classification, TBT concentration was apparently unaffected by trophic level. In contrast, TPT concentration was higher in organisms belonging to higher trophic levels in the deep water, suggesting the bioconcentration of TPT through the food web.

Takahashi et al. (1997b) have previously proposed the concentration of BTs along food chains. Bigger carnivorous fish and scavengers that are at the higher trophic levels of the food web contained relatively elevated levels of BT residues in their tissues. The bio magnification factor (BMF tissues relative to their stomach contents) was estimated roughly for BTs. These values ranged from 0.8 (in dories) to 1.8 (in dogfish sharks). Although the increased exposure by feeding on contaminated diet would be expected, generally, to result in accumulation in higher trophic species, BMFs for BTs were estimated to be less than 10 in most species including deep-sea fish. This may be due to the biodegradable nature of TBT in higher organisms, and/or lower assimilative potency as compared to other persistent contaminants such as PCBs and methylmercury.

TBT was the predominant OTs compounds in almost all deep sea biota (Takahashi et al. 2001). This suggests a continuing input of TBT into the deep-sea environment and a relatively low capacity to degrade TBT in these organisms.

Carbon and nitrogen isotopes are useful to characterize the trophic level of an organism (Broman et al. 1992). Iwasaki et al. 2005 (personal communication) has classified various species of deep-sea organisms by $\delta^{13}C$ value into two groups. Group A includes *C. nankaienssi*, *Cl. tsubasa*, and *Cl. nautilei* and group B *P. verrucosa*, *M. albatrossae*, and *M. subsquamosa*. The organisms in group A use the organic matter chemosynthesized by symbiotic bacteria as nutrient source, while those in group B depend on photosynthesis carried out near the surface by phytoplankton. The relationship between $\delta^{13}C$ and OT concentration is shown in Fig. 6.5. No difference in BT or TBT concentration was observed between the two groups, but PT and TPT concentrations were significantly higher in group B than in group A ($p < 0.001$). Trophic levels in the food chain are estimated using $\delta^{15}N$ values (Broman et al. 1992). The relationship between $\delta^{15}N$ and OT concentration is shown in Fig. 6.6. No change in BT or TBT concentration was observed to accompany the increase of $\delta^{15}N$ values, across trophic levels, but PT and TPT concentration generally increased with increasing of $\delta^{15}N$ values. These findings suggested that TPT concentration rises as trophic level increases, but no such trend is observed for TBT.

The partition coefficient of OT compounds between deep-sea organisms and sediment, were calculated by dividing the TBT concentration in each deep-sea organism by the concentration in sediment. The average partition coefficients of TBT for *C. nankaiensis*, *P. verrucosa*, *M. albatrossae*, *M. subsquamosa*, *Cl. tsubasa* and *Cl. nautilei* were 2.6, 0.72, 0.63, 0.19, 0.43, 0.30 and 0.46, respectively. *C. nankaiensis* is a carrion feeder as well as a deposit feeder. The high partition coefficient for *C. nankaiensis* may therefore be due to bioavailability if TBT in its prey.

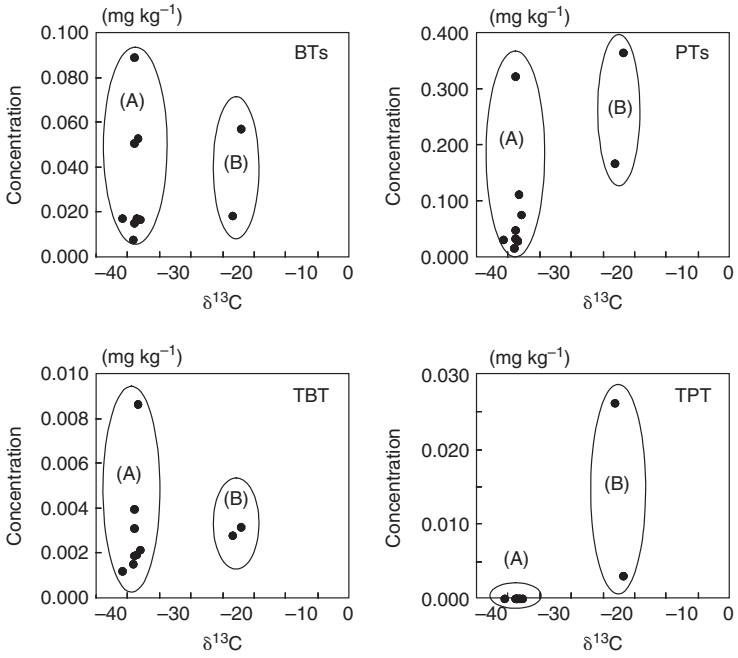

Fig. 6.5 The relationship between BTs or PTs and $\delta^{13}C$

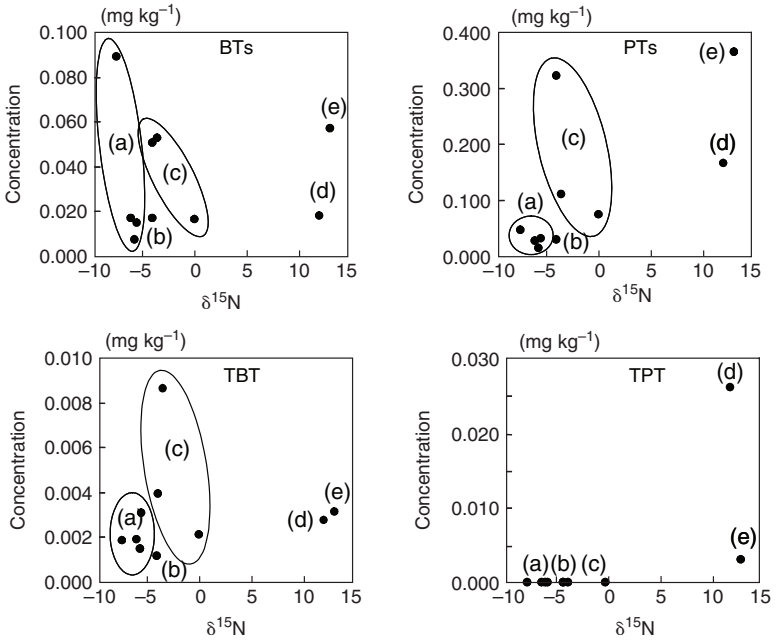

Fig. 6.6 The relationship between BTs or PTs and $\delta^{13}N$

6.5 Transportation of Organotin Compounds to Deep Sea Area

The transportation process whereby harmful chemical compounds reach deep sea environments is of great concern. Many artificial chemical compounds such as organochlorine insecticide residues have been detected in the deep sea environment. However, about the process behind transport of these chemicals are uncertain. In the case of TBT, the degradation rate following release into water from vessel hulls is relatively fast (half-lives; 7–19 days). TBT is thus less persistent in the aquatic environment than many organochlorines. Nevertheless, BT and PT residues have been detected in deep-sea organisms and sediments. Various hypotheses have been proposed concerning the process of OT transportation to bathyal regions. Michel and Averty (1999) estimated the vertical transport of OTs in Mediterranean Sea. The fraction of TBT absorbed by suspended particulate material (SPM) is less than 10^{-3}, because Mediterranean Sea waters are not very turbid: the mean SPM is of the order of 0.28 mg l^{-1} and particulate carbon content is between 0.024 and 0.036 mg l^{-1} for waters below 200 m. Furthermore, the sedimentation rate on the highest abyssal plains in 0.01 cm $year^{-1}$. Considering the scientific data, it is hard to imagine that TBT which adheres to SS will deposited to deep sea environment in significant quantities. Dissolved TBT may, however, have been transported vertically down to the 1,200 m level. The presence of TBT in deep waters in this region is therefore more likely to be due to vertical movements of water masses of surface origin.

Harino et al. (1998) reported that OTs are concentrated or absorbed readily on suspended substances such as plankton and OTs which are absorbed to sediment and suspended substance, may persist for a considerable period (Dowson et al. 1993). Where present in significant densities, such suspended material may be responsible for transport and deposition of absorbed OTs in the bathyal region. OTs also accumulate in biota such as marine mammals and fish and the bioconcentration factor for these biota are reported to be very high (Le et al. 1999; Yamada and Takayanagi 1992; Harino et al. 1998, 2000): The carcasses of these organisms are also deposited in the bathyal region and represent a further transport route.

6.6 Conclusions

As the deep sea is remote from human activity, it has been assumed that contamination in the bathyal region will not have an influence on our lives, even if polluted. Therefore, in the past, parts of the deep sea have been used for disposal of waste. However, this is a false assumption. The deep sea has a close relationship with human life. Recently, deep sea water has been sold as for consumption and deep sea fish are being taken, increasingly, as food. Furthermore, the deep sea may have an influence on the coastal environment by determining ocean currents and transportation of aquatic organisms. With the development of the submersible research

vessels, the pollution situation of the deep sea is beginning to be unravelled. It is regrettable that the deep sea environment has been found to be polluted by OT compounds, especially, TPT which was banned in Japan at an early stage. In order to help preserve the deep sea environment, the top research priority is to elucidate the transport process of OTs. The transportation process of OTs into deep sea environment is largely unknown at present, although some hypotheses have been proposed. Furthermore, it is also important to survey the persistence of these compounds in the bathyal region. Clearly there are a number of unresolved issues surrounding, the fate of OT contamination in deep sea environment.

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