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Organochlorine and Butyltin Residues in Mesopelagic Myctophid Fishes from the Western North Pacific

SHIN TAKAHASHI,† SHINSUKE TANABE,*,† AND KOUICHI KAWAGUCHI‡

Center for Marine Environmental Studies, Ehime University, Tarumi 3-5-7, Matsuyama 790-8566, Japan, and Ocean Research Institute, The University of Tokyo, Minamidai 1-15-1, Nakano, Tokyo 164, Japan

Organochlorine (OCs) and butyltin (BTs) residues were determined in mesopelagic myctophid fishes collected from the western North Pacific to elucidate contamination status, accumulation patterns, and distribution. Among OCs, concentrations of PCBs (polychlorinated biphenyls) and DDTs (DDT and its metabolites) were the highest in myctophids (at maximum concentrations of 370 ng/g and 280 ng/g lipid wt, respectively). CHLs (chlordane compounds) or HCHs (hexachlorocyclohexanes) were the next most abundant OCs, and HCB (hexachlorobenzene) was the lowest. The maximum concentration of total butyltin (∑BTs = MBT + DBT + TBT) in fish was 46 ng/g wet wt. Concentrations of PCBs, CHLs, and BTs were significantly lower in oceanic myctophids than those in fishes from Japanese coastal water, Suruga Bay. The residue pattern of OCs and BTs showed a specific trend according to diel vertical migration of myctophids. Relatively high concentrations of PCBs, DDTs, and CHLs were found in nonmigratory species living in deeper waters, whereas concentrations of HCHs, HCB, and BTs were high in migratory species, which migrate up to the upper 200 m at night for feeding. These patterns are also influenced by the vertical distributions of OCs and BTs in the subarctic and transitional waters of the western North Pacific, where intrusion of watermass from the Okhotsk Sea has been suggested. Temporal variation in the residue level and composition of OCs found in myctophids indicated a significant decrease in the input of DDTs into the marine environment while HCHs declined at a slower rate.

Introduction

During the last few decades, pollution by persistent man-made chemicals such as organochlorines (OCs) has spread all over the world as evidenced by their detection in various environmental components and biota including those far from human activities (*1*−*9*). In this regard, it has been emphasized in several studies that deep-sea sediments play a role as a sink and final reservoir for persistent contaminants (*10*−*13*). During the 1970s and 1980s, several monitoring studies have reported the accumulation of OCs in deep-sea organisms collected from the Atlantic Ocean and around the U.S. coasts (*14*−*20*). Despite this, only a few studies have been conducted on contamination by OCs in the deep-sea environment in recent years (*21, 22*). In particular, little data are available in the western North Pacific region, except in the marginal Japan Sea (*23*) and Suruga Bay (*24*).

In addition to OC contamination, aquatic pollution by butyltins (BTs), particularly toxic tributyltin (TBT) used as a biocide in antifouling paints for boats and aquaculture nets, has been of great concern due to their bioaccumulative potential and deleterious effects in organisms (reviewed in refs *25* and *26*). Although a reduction in TBT contamination was recorded after the restriction on TBT usage implemented in the most developed countries since the late 1980s, TBT concentrations in the coastal waters of several regions still persist at levels toxic to susceptible organisms such as gastropods (*27*−*29*). Furthermore, considerable concentrations of BTs were detected in higher trophic aquatic organisms such as cetaceans, pinnipeds, and seabirds from the North Pacific and Asian coastal waters (*30*−*35*). These findings suggest possible long-term contamination and toxic threat of BTs in the global marine ecosystem. However, the detection of BTs in deep-sea organisms has been scarcely reported (*36*).

Our earlier studies were conducted in Suruga Bay, which is a deep coastal body of water along the Pacific Coast of Japan (*24, 36, 37*). In these studies, accumulation of OCs and BTs was reported in all deep-sea organisms, suggesting their widespread contamination even in the deep-sea ecosystem. However, contamination of these compounds in deep-sea ecosystem of the Pacific Ocean waters is still unclear. Therefore, the present study determined polychlorinated biphenyls (PCBs), DDT and its metabolites (DDTs), chlordane compounds (CHLs), hexachlorocyclohexane isomers (HCHs), hexachlorobenzene (HCB), and BTs including TBT and its degradation products, di- (DBT) and monobutyltin (MBT) in myctophid fishes (family: Myctophidae) in the mesopelagic zone of the western North Pacific Ocean.

The fishes belonging to the family Myctophidae (including about 250 species) are distributed widely in the world oceans and show the species-specific diel vertical migration patterns in the water column (*38*). Such characteristics of myctophids can be useful for monitoring contaminants in the open waters. In the present study, concentrations of OCs and BTs in myctophids were compared with those of the coastal fish reported in earlier studies to elucidate horizontal distribution and temporal trends in contamination. In addition, interspecies comparison of the accumulation profiles of OCs and BTs in relation to the diel vertical migration modes of each myctophid species were examined to delineate vertical distribution of the contaminants in the western North Pacific.

Materials and Methods

Samples. Six species of mesopelagic myctophids were collected from 50−100 m (nighttime) and 200−700 m depth (daytime) at four locations in the western North Pacific (38−41° N, 143−147° E) in 1994 (Table 1 and Figure 1). Myctophid fishes undergo species-specific diel vertical migration between mesopelagic (200−1000 m) and epipelagic (<100 m) zones. Figure 2 shows the four modes of vertical migration of myctophid species: (i) nonmigratory species, *Stenobrachius nannochir* and *Lampanyctus regalis*, stay in deep water throughout day and night; (ii) migratory species, *Diaphus theta* and *Ceratoscopelus warmingi*, migrate up to shallow water for feeding at night; (iii) semi-migratory species, *Stenobrachius laucopsarus*, a part of population migrate to shallow water at night, while the remainder do not migrate;

* Corresponding author fax: +81-89-946-9904; e-mail: shinsuke@agr.ehime-u.ac.jp.
† Ehime University.
‡ The University of Tokyo.

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TABLE 1. Sampling Data and Measurements of Myctophid Species Collected from the Western North Pacific

species	location, date, and depth	*n*	mean length[a] (cm)	mean wt (g)	diel migration[b]
Stenobrachius leucopsarus	station A, 08/24/94 night, 500−600 m	4	9.3 (8.7)	7.5	semi-migrant (1)
Lampanyctus jordani	station A, 08/24/94 night, 500−600 m	14	13.2 (11.7)	18.4	semi-migrant (2)
Lampanyctus regalis	station A, 08/24/94 night, 500−600 m	2	19.5 (17.2)[c]	58.1	nonmigrant
Stenobrachius nannochir	station A, 08/24/94 night, 500−600 m	4	11.4 (9.9)	10.7	nonmigrant
Stenobrachius leucopsarus	station B, 08/18/94 day, 400−500 m	7	9.4 (8.0)	7.8	semi-migrant (1)
Ceratoscopelus warmingi	station B, 08/18/94 day, 400−500 m	14	9.4 (8.5)	7.7	migrant
Diaphus theta	station B, 08/18/94 day, 400−500 m	30	9.4 (8.2)	7.8	migrant
Lampanyctus regalis	station B, 08/18/94 day, 400−500 m	1	18.2 (16.5)	41.8	nonmigrant
Ceratoscopelus warmingi	station C, 08/19/94 night, 50−100 m	33	9.1 (7.9)	5.8	migrant
Stenobrachius nannochir	station D, 08/08/94 night, 500−600 m	24	10.9 (9.6)	8.4	nonmigrant

[a] Total length (fork length in parentheses). [b] See Figure 2. [c] Adults stop diel vertical migration.

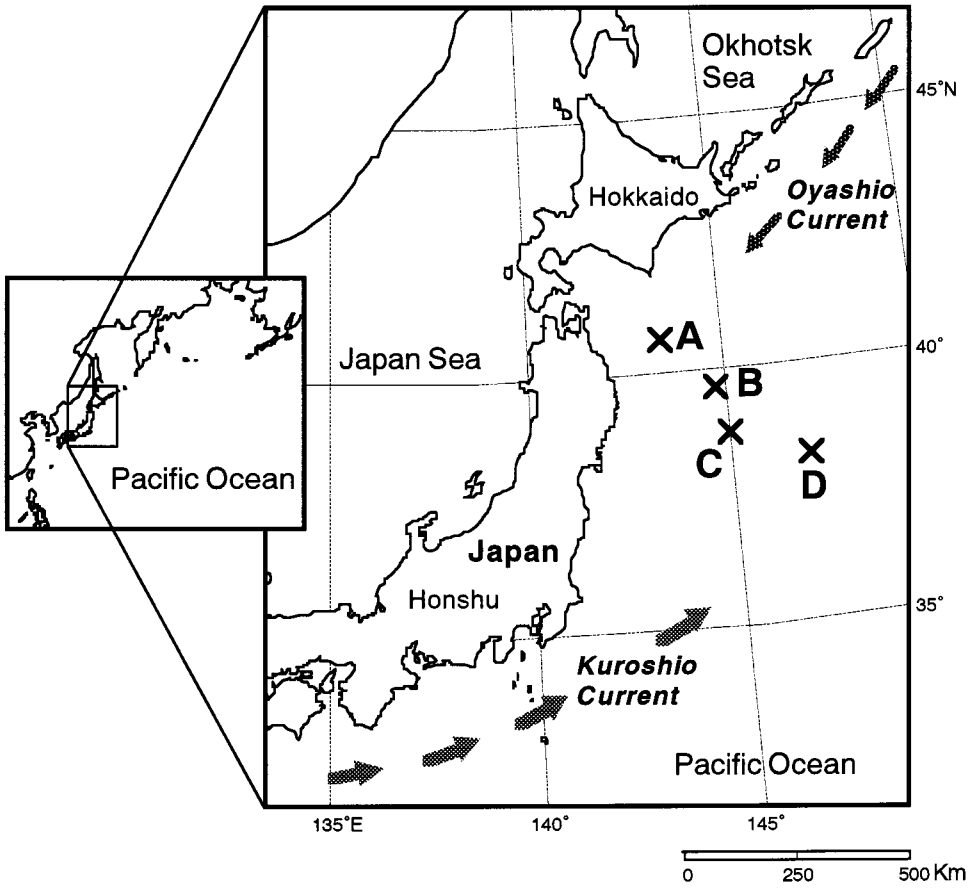

FIGURE 1. Map showing the sampling locations (×) in the western North Pacific (station A: 40°59.3′ N, 143°24.6′ E; station B: 39°46.4′ N, 144°59.3′ E; station C: 38°59.9′ N, 145°00.5′ E; station D: 38°11.9′ N, 147°01.6′ E). Arrows in the figure indicate major ocean currents along the Pacific Coast of Japan.

(iv) *Lampanyctus jordani*, the upper limit of vertical distribution is shifted to shallower depth at night (*39*). The details of biometry of myctophids investigated are shown in Table 1. The whole body of individual specimens belonging to the same species collected from the same sampling location was pooled and homogenized to prepare a composite sample, which was employed for chemical analysis.

Chemical Analysis. Organochlorine pesticides, DDTs (*p*,*p*′-DDE, *p*,*p*′-DDD, and *p*,*p*′-DDT), CHLs (*cis*-chlordane, *trans*-chlordane, *cis*-nonachlor, *trans*-nonachlor, and oxychlordane), HCHs (α, β, and γ isomers), HCB, and PCBs were analyzed following the method described by Tanabe et al. (*40*). Briefly, a sample was homogenized with anhydrous Na_2SO_4 and extracted using a Soxhlet apparatus with a mixture of diethyl ether and hexane. Lipid content was determined from an aliquot of the extract. The extract was then added into a dry Florisil column to remove fat and eluted with 20% water−acetonitrile. OCs in the eluate were transferred to hexane. After concentration, the hexane extract was cleaned with sulfuric acid and separated into two fractions with Florisil packed into a glass column. The first fraction eluted with hexane contained PCBs, *trans*-nonachlor, *p*,*p*′-DDE, and HCB. The second fraction eluted 20% dichloromethane in hexane contained other OC pesticides.

Sample extracts were analyzed by capillary gas chromatography with a ^{63}Ni electron capture detector (GC-ECD). Chromatographic separation was performed on a Hewlett-Packard 5890 series II gas chromatograph with a 30 m × 0.25 mm (i.d.) DB-1 capillary column coated at 0.25 μm film thickness (J&W Scientific Co., Folsom, CA, 100% dimethyl−polysiloxane). The concentrations of organochlorine insecticides were quantified from individually resolved peak areas with the corresponding peak areas of authentic standards. For the quantification of PCBs, an equivalent mixture of

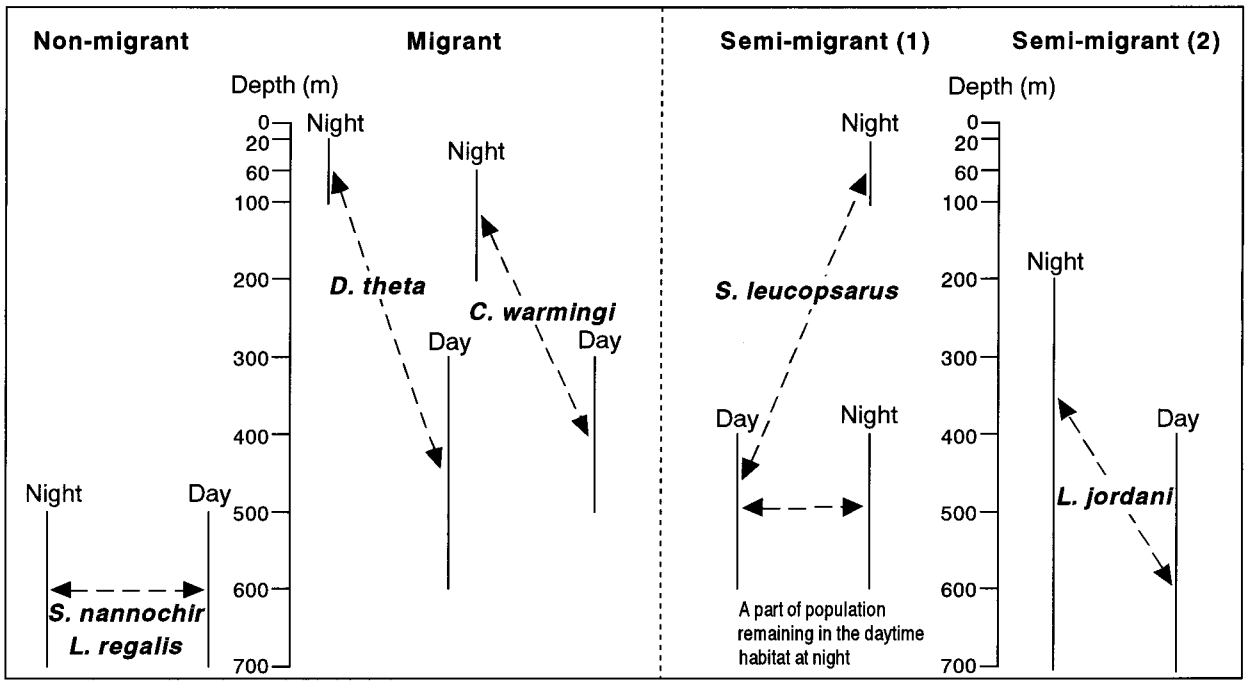

FIGURE 2. Diel vertical migration patterns of myctophid fishes in the subarctic waters of the western North Pacific. Semi-migrants (1) migrate to the upper 100 m during night, but a portion of the population remains as they were during the daytime, showing bimodal vertical distribution pattern. Semi-migrants (2) with the upper limit of depth distribution shifts to shallower layer at night than daytime habitat but rarely occur in the upper 100 m layer.

Kanechlor 300, 400, 500, and 600 was used as a standard. Total PCB concentration was calculated by adding the concentrations of individual resolved peaks. Peak identification (101 peaks of PCBs including 117 major congeners and isomers) was reported previously (*41*). The recoveries in this analytical procedure were 100 ± 13% for insecticides (average of all OC pesticide descried above) and 102 ± 15% for total PCBs ($n = 4$). Quality assurance for the measurement of OCs in our present technique was confirmed by analyzing Standard Reference Material 1945 (Organics in Whale Blubber) provided from The National Institute of Standards and Technology (NIST), and the results agreed well with the NIST certified values. Concentrations of OCs were not corrected for the recoveries and are reported as nanograms per gram on a lipid weight basis.

BTs were analyzed following the method described by Takahashi et al. (*42*). Briefly, a sample was homogenized with 0.1% tropolone−acetone and 2 N HCl. BTs in the extract were transferred to 0.1% tropolone−benzene, and the moisture in the solvent was removed with anhydrous Na_2SO_4. BTs in the extract were then propylated by adding *n*-propylmagnesium bromide as a Grignard reagent. After decomposition of the excess Grignard reagent with 1 N H_2SO_4, the derivatized extract was transferred to10% benzene−hexane. The extract was then passed through Florisil-packed glass column (eluting with hexane). The final hexane eluate was concentrated and subjected to GC quantification.

Sample extracts were analyzed by capillary gas chromatography with a flame photometric detection (GC−FPD). Chromatographic separation was performed on a Hewlett-Packard 5890 series II gas chromatograph with a 30 m × 0.25 mm (i.d.) DB-1 capillary column. The flame photometer was equipped with a 610-nm band-pass filter that is selective for tin-containing compounds. Monobutyltin trichloride, dibutyltin dichloride, and tributyltin chloride of known amounts (0.1 μg each) spiked into the liver of Antarctic minke whale containing undetectable levels of butyltin residues was concurrently run with samples through the whole analytical procedure for use as an external standard. Procedural blanks were included with every batch of sample analysis. Detection limits (assigned twice the values of the blanks) of MBT, DBT, and TBT in tissues were 5.0, 2.0, and 5.0 ng/g wet wt, respectively. The recovery rates through the whole analytical procedure for MBT, DBT, and TBT spiked into the liver of Antarctic minke whale were 104 ± 19.6%, 117 ± 14.3%, 108 ± 5.2%, respectively ($n = 4$). In addition, hexyl tributyltin was added as an internal standard, and its recoveries through the analytical procedure were more than 80%. The concentrations of BTs were not corrected for the recovery and are reported as nanograms of corresponding ion (MBT^+, DBT^+, or TBT^+) per gram on a wet weight basis in this study.

Results and Discussion

Contamination Status and Distribution of OCs. Organochlorine compounds (OCs) were detected in all mesopelagic myctophids collected from the western North Pacific off northern Japan. Among the OCs analyzed, concentrations of PCBs and DDTs (up to 370 and 280 ng/g lipid wt, respectively) were the highest, and those of other OCs were approximately in the order of CHLs ≥ HCHs > HCB (Table 2). While concentrations of OCs varied among the species, no significant differences in the concentrations of fishes were noted between station A and station B ($p > 0.05$, Mann−Whitney *U*-test). In addition, concentrations of OCs in both species from stations C (*Ceratoscopelus warmingi*) and D (*Stenobrachius nannochir*) were similar to those of species from stations B and A, respectively (Table 2). This suggests no apparent spatial difference in the contamination among the sampling locations. The predominance of PCBs and DDTs in myctophids has been similar to those in fish and marine mammals collected from Japanese coastal waters and the western North Pacific regions (*24, 31, 40, 43, 44*). This is due to higher bioaccumulative properties of these compounds and continuous release of PCBs into the environment.

Compared to deep-sea fishes from Japanese coastal water, Suruga Bay (*24*), the concentrations of PCBs and CHLs in myctophids from open ocean were apparently lower ($p < 0.01$, Mann−Whitney *U*-test). Concentrations of DDTs and

TABLE 2. Concentrations of Organochlorine Pesticides and PCBs (ng/g Lipid wt) in Mesopelagic Myctophids Collected from the Western North Pacific

samples	lipid (%)	*p,p′*-DDE	*p,p′*-DDD	*p,p′*-DDT	∑DDT[a]	α-HCH	*β*-HCH	*γ*-HCH	∑HCH[b]
				Station A					
Stenobrachius leucopsarus	17	21	5.5	15	42	15	6.8	2.2	24
Lampanyctus jordani	9.4	61	10	30	110	15	3.8	2.3	21
Lampanyctus regalis	13	150	3.2	120	270	6.0	2.2	0.86	9.1
Stenobrachius nannochir	18	45	14	62	120	7.3	3.0	1.0	11
				Station B					
Stenobrachius leucopsarus	18	24	5.6	14	44	8.5	4.6	1.3	14
Ceratoscopelus warmingi	23	11	3.7	8.3	23	3.5	4.6	1.2	9.3
Diaphus theta	4.9	71	23	62	160	13	4.5	2.0	20
Lampanyctus regalis	17	120	21	140	280	1.4	1.4	0.25	3.1
				Station C					
Ceratoscopelus warmingi	22	7.4	2.4	5.1	15	7.7	3.2	1.1	12
				Station D					
Stenobrachius nannochir	20	33	9.0	37	79	2.8	2.2	0.50	5.5

samples	oxychlordane	*trans*-chlordane	*cis*-chlordane	*trans*-nonachlor	*cis*-nonachlor	∑CHL[c]	HCB	PCBs
			Station A					
Stenobrachius leucopsarus	0.91	2.2	3.6	6.7	1.3	15	5.1	68
Lampanyctus jordani	1.2	2.4	6.6	12	2.2	24	8.0	200
Lampanyctus regalis	1.7	3.5	7.8	19	2.6	35	3.6	370
Stenobrachius nannochir	1.6	4.8	9.5	9.8	2.5	28	1.3	110
			Station B					
Stenobrachius leucopsarus	0.70	1.8	4.1	6.3	1.2	14	3.9	110
Ceratoscopelus warmingi	0.43	1.3	2.4	1.6	0.86	6.6	3.3	49
Diaphus theta	2.3	5.9	15	18	5.7	47	7.9	240
Lampanyctus regalis	2.0	2.2	7.1	17	4.3	33	0.79	280
			Station C					
Ceratoscopelus warmingi	0.19	0.76	1.6	1.8	0.64	5.0	2.4	20
			Station D					
Stenobrachius nannochir	0.88	3.0	5.7	6.0	1.5	17	0.80	130

[a] ∑DDT = *p,p′*-DDE + *p,p′*-DDD + *p,p′*-DDT. [b] ∑HCH = α-HCH + *β*-HCH + *γ*-HCH. [c] ∑CHL = oxychlordane + *trans*-chlordane + *cis*-chlordane + *trans*-nonachlor + *cis*-nonachlor.

HCHs also seemed to be lower in mesopelagic myctophids; however, statistical analysis showed less significant difference in the concentrations between the fishes ($p = 0.073$ and 0.039, respectively). These results would reflect the horizontal distribution of OCs in the western North Pacific and the elevated contamination of PCBs and CHLs around Japanese coastal waters. It has been noted that PCBs used in electrical equipments are continuously released into the environment (*45*). In Japan, CHLs had been used largely as a termiticide until 1986, whereas DDT and technical HCH were banned in the early 1970s. In an ocean monitoring study using squids (*46*), higher concentrations of PCBs were also found in squids collected around Japanese coastal waters.

Concentrations of OCs in mesopelagic myctophids analyzed in this study were lower than those in deep-sea organisms collected from the Atlantic Ocean (*14*−*18*), Gulf of Mexico (*16*), U.S. coasts (*19*, *20*), and the Arctic Ocean (*5*,*21*) (Table 3). This suggests a relatively low contamination of OCs in the western North Pacific as compared to other regions. In addition to this, a decreasing trend of OC inputs into the marine environment during the last few decades may lead to lower concentrations in myctophids collected in 1994; most data presented in Table 3 were collected from deep-sea samples collected in the 1970s and 1980s. However, variations in analytical methods and fish species compared in Table 3 may influence the different levels of OCs. Comparison of myctophid fishes analyzed by a uniform technique is required to provide a better understanding on these questions.

Concentrations of PCBs, DDTs, and HCHs in myctophid species, *D. suborbitalis*, collected from Yaizu, Suruga Bay, in 1976 were 1500, 1300, and 69 ng/g lipid wt, respectively (*47*), which are higher than those in myctophids collected in 1994 by 1 order of magnitude for PCBs and DDTs and by a factor of 5 for HCHs. While all the species of myctophids analyzed in this study are offshore species, *D. suborbitalis* is a coastal species found along Suruga Bay (*48*). Thus, the difference in the concentrations of PCBs between these offshore and coastal myctophids are expected to be due not only to a temporal decline but also to spatial variation, particularly in the case of PCB concentrations as suggested above. The earlier studies on the temporal trends of OCs in northern fur seals from the Pacific Coast of Japan (*40*) and minke whales from the western North Pacific (*8*) have also reported a significant decline in the concentrations of DDTs, but smaller rates of reductions were observed in the concentrations of PCBs and HCHs from the 1970s to 1990s. A similar observation is found in the present study on myctophids.

Contamination Status and Distribution of BTs. BTs were also detected in almost all myctophids analyzed except for *C. warmingi* from station B and *S. nannochir* from station A (Table 4). The highest concentrations of total butyltins (∑BTs: MBT + DBT + TBT) were up to 46 ng/g wet wt. To our knowledge, this is the first report of detection of BTs in deep-sea organisms from the mesopelagic zone. This result suggests the expansion of butyltin contamination on a global scale. As discussed for OCs, concentrations of BTs varied among the species, but did not differ significantly among the sampling locations ($p > 0.05$; the data of all fish samples from stations A and B were employed for Mann−Whitney *U*-test).

The concentrations of BTs in myctophids were comparable to those in squids from open waters of the western North Pacific (*46*). However, these concentrations were lower

TABLE 3. Mean Concentrations of PCBs and DDTs in Deep-Sea Organisms from Various Parts of the World

species	year	location	depth (m)	PCBs ng/g wet	PCBs ng/g lipid wt	DDTs ng/g wet	DDTs ng/g lipid wt	ref
Chauliodontidae	1970−72	North & South Atlantic	450−900	26[a]	3000[a]	6.2[b]	760[b]	15
Stenophoridae	1972	North & South Atlantic	130−660	33[a]	2200[a]	8.3[b]	480[b]	15
Gonostanomiae	1972	North & South Atlantic	510−660	79[a]	15000[a]	14[b]	4300[b]	15
blue hake	1972	Atlantic (Cape Hatteras)	2500	–	–[c]	5400[d]	–	14
(*Antimora rostrata*)	1972−74	Atlantic (Cape Hatteras)	2500	–	–	6900[d]	–	17
Mesopelagic fish	1973	Gulf of Mexico	0−760	200[a]	13000[a]	19[b]	2000[b]	16
black scabbord (*Aphanopus carbo*)		Atlantic (Madeira Island)	800−1200	–	5800[e]	–	9200[b]	18
myctophids (*Diaphus suborbitalis*)	1984	Suruga Bay	–	48[i]	1500[i]	43[b]	1300[b]	47
rattail		Hudson Canyon	3200	2700[f]	–	–	–	19
(*Coryphaenoides armatus*)		Carson Canyon	1700−2300	360[f]	–	–	–	19
amphipods (*Eurythenes gryllus*)	1983	Arctic Ocean	2075	–	22000[a]	–	13000[b]	5
glacial ellpout (*Lycodes frigidus*)	1983	Arctic Ocean	2075	–	2300[a]	–	1500[b]	5
Dover sole	1985	Farallon Island	500	830[g]	11000[g]	760[b]	11000[b]	20
(*Microstomus pacificus*)	1985	off California	1000	980[g]	12000[g]	2300[b]	22000[b]	20
sable fish (*Anoplopoma fimbria*)	1985	off California	1000	7000[g]	56000[g]	8700[b]	74000[b]	20
cartilaginous fish (2 sp.)	1992	Davis Strait (Greenland)	800−2200	310[h]	430[h]	490[b]	690[b]	21
bony fish (6 sp.)	1992	Davis Strait (Greenland)	200−2100	300[h]	790[h]	250[b]	670[b]	21
fish (6 sp.)	1993−94	Suruga Bay, Japan	200−740	160[i]	910[i]	60[b]	360[b]	24
crustaceans (9 sp.)	1993−94	Suruga Bay, Japan	180−980	73[i]	1200[i]	26[b]	430[b]	24
cephalopods (3 sp.)	1993−94	Suruga Bay, Japan	135−540	79[i]	1300[i]	37[b]	620[b]	24
echinoderms (4 sp.)	1993−94	Suruga Bay, Japan	310−980	22[i]	1300[i]	3.5[b]	210[b]	24
myctophids (6 sp.)	1994	Western North Pacific	50−600	20[i]	160[i]	16[b]	110[b]	this study

[a] Total PCB concentrations determined as Aroclor equivalent (Aroclor 1254 used as a standard). [b] Sum of *p,p′*-DDE, *p,p′*-DDD, and *p,p′*-DDT. [c] –, no data available. [d] Sum of *p,p′*-DDE, *p,p′*-DDD, *p,p′*-DDT, and *o,p′*-DDT. [e] Sum of 6 PCB congeners (IUPAC Nos. 28, 52, 101, 128, 158, and 180). [f] Sum of 24 PCB congeners (IUPAC Nos. 28, 44, 49, 52, 60, 70, 86, 87, 95, 101, 105, 128, 129, 137, 138, 141, 153, 156, 180, 183, 194, 195, 205, 206, and 207). [g] Total PCB concentrations determined as Aroclor equivalent (mixture of Aroclor 1242, 1254, and1260 used as a standard). [h] Sum of 19 PCB congeners (IUPAC Nos. 28, 74, 99, 101, 105, 110, 118, 128, 138, 141, 153, 156, 157, 170, 180, 187, 194, 206, and 209). [i] Total PCB concentrations determined as Kanechlor equivalent (mixture of Kanechlor 300, 400, 500, and 600 used as a standard).

TABLE 4. Concentrations of Butyltin Compounds (ng/g wet wt) in Mesopelagic Myctophids Collected from the Western North Pacific

samples	moisture content (%)	MBT	DBT	TBT	∑BT[a]	TBT/∑BT (%)
		Station A				
Stenobrachius leucopsarus	65	<5.0	11	20	31	65
Lampanyctus jordani	82	12	4.3	30	46	65
Lampanyctus regalis	68	9.0	<2.0	8.9	18	49
Stenobrachius nannochir	64	<5.0	<2.0	<5.0	ND[b]	NA[c]
		Station B				
Stenobrachius leucopsarus	70	9.2	1.9	15	26	58
Ceratoscopelus warmingi	63	<5.0	<2.0	<5.0	ND	NA
Diaphus theta	76	7.1	4.1	35	46	76
Lampanyctus regalis	67	<5.0	4.9	7.4	12	62
		Station C				
Ceratoscopelus warmingi	61	10	<2.0	<5.0	10	NA
		Station D				
Stenobrachius nannochir	67	7.6	<2.0	5.3	13	41

[a] ∑BT = MBT + DBT + TBT. [b] ND, butyltin compounds were not detected. [c] NA, not available.

than those in shallow-water fish from Japanese coastal waters (*33*, *36*, *43*, *49*, *50*) and from various coastal regions of the United States (*51*, *52*) and European countries (*53*−*56*). Elevated contamination by BTs around Japanese coastal waters was also suggested in recent monitoring studies using squids (*46*) and marine mammals (*35*). Furthermore, the concentrations of BTs in myctophids analyzed in this study were lower than those in deep-sea fishes from Suruga Bay ($p < 0.05$, Mann−Whitney *U*-test), and the concentrations of both deep-sea fishes were significantly lower ($p < 0.01$) than those of shallow-water fishes from Suruga Bay (*36*). These observations suggest that the areas highly contaminated by BTs are localized in coastal shallower waters, which may be due to the fresh and continuous input of BTs into the coastal-surface regions through human activities and the lower mobility of BTs.

Accumulation Patterns of OCs and BTs in Relation to Migration Patterns of Fishes. To discuss the accumulation patterns of OCs and BTs in relation to the migration patterns, the concentrations of OCs and BTs were compared among the various migratory types of fish. The accumulation patterns of OCs and BTs showed a specific trend in accordance with the migration types (Figure 3). Except for a semi-migrant species *D. theta*, higher concentrations of PCBs, DDTs, and CHLs were found in non- or less-migratory species living in deeper waters (Figure 3a). On the contrary, HCHs, HCB, and BTs were accumulated at higher concentrations in migratory species migrating to shallower waters as compared with nonmigratory fishes (Figure 3b). Except for *L. regalis* having apparently larger body size than the other species, myctophids analyzed in this study have similar body size and feed mainly on copepods (*38*). Thus, most of the fishes can

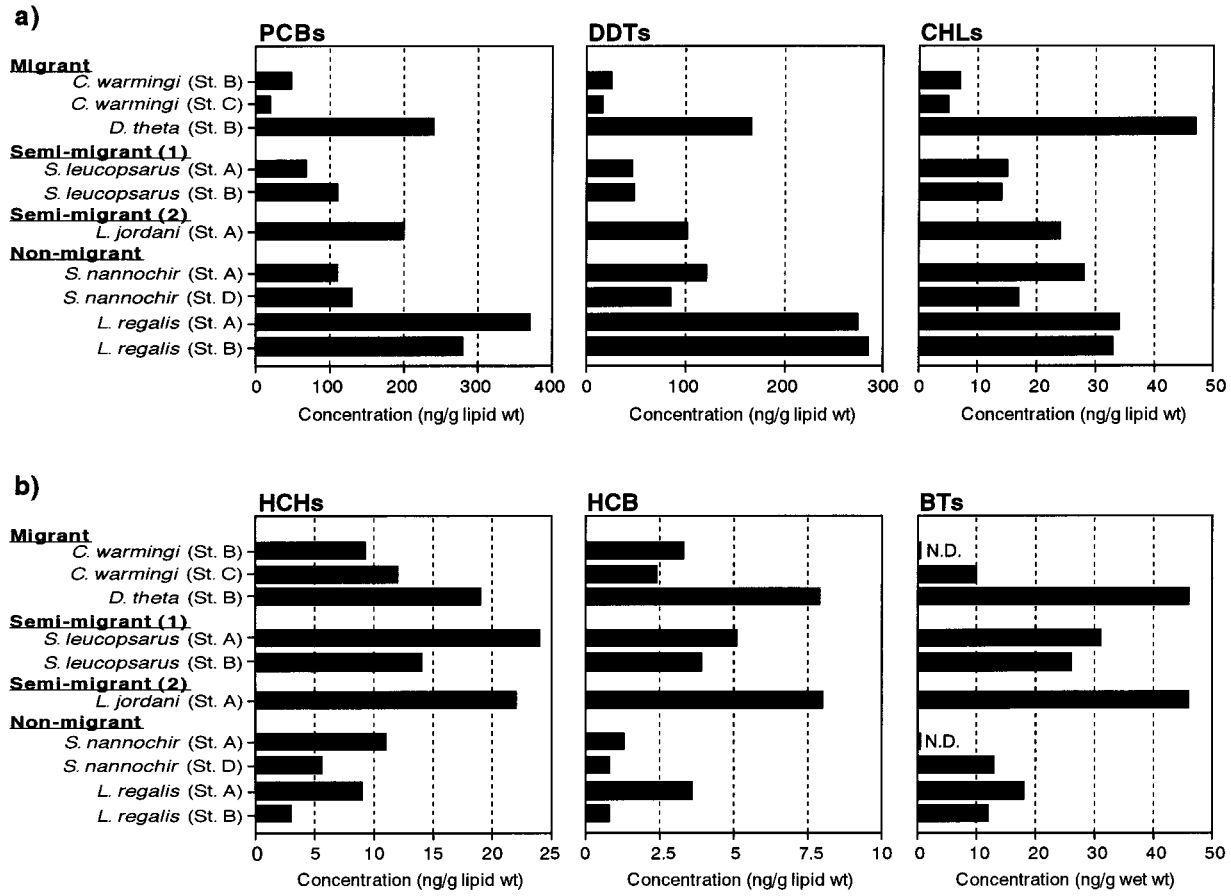

FIGURE 3. Concentrations and residue patterns of PCBs, DDTs, CHLs, HCHs, HCB, and BTs in myctophid fishes with different migration types. The residue patterns were grouped into (a) higher concentrations of PCBs, DDTs, and CHLs observed in species living in deeper waters and (b) higher concentrations of HCHs, HCB, and BTs observed in species migrating to shallower waters.

be expected to be in the same trophic level. Only *L. regalis* often feed squids, shrimps and mysids, and thus this species can be ranked to be at higher trophic level. This may be a reason for higher concentrations of some organochlorine compounds observed in *L. regalis*. However, as shown in Figure 3a,b, results cannot be fully explained by the differences in trophic level in myctophid species.

Tanabe et al. (*57*) showed that the concentrations of HCHs significantly decreased with depth in the open water column of the Pacific Ocean because of their volatility and high atmospheric mobility. In the Arctic Ocean, HCHs and HCB, which are volatile OCs with lower molecular weight, were found at higher concentrations in the surface waters (*4*). Thus, the accumulation pattern found in myctophids seems to reflect such vertical distribution of HCHs and HCB in the water column. On the other hand, the profiles of PCBs and DDTs found in this study were not consistent with the vertical distribution reported by Tanabe et al. (*57*); rather a uniform vertical distribution of PCBs and DDTs were reported by Tanabe et al. (*57*) for waters collected in 1979. It can be expected that the vertical profiles of PCBs and DDTs in the water column have changed following the recent reduction of their input into surface waters. This hypothesis can be supported by more recent investigation on the vertical profile of PCBs in the open-water column of the Japan Sea, which reported a lower concentrations of dissolved PCBs in the surface water and maximum concentration at a depth of 500 m (*23*). Hargrave et al. (*5*) reported that benthic amphipods from the Arctic continental shelf contained between 10 and 60 times the concentrations of PCBs, DDTs, and CHLs but only 2−5 times those of HCHs and HCB found in pelagic plankton and suggested that OCs with lower water solubility and higher affinity for absorption to particles should be transported to depth and incorporated into the marine food web more readily than more water-soluble compounds. Accumulation profiles of these compounds found in the myctophids (Figure 3) also supported such a hypothesis. Thus, organisms living in deep waters may be at a risk of exposure to higher molecular weight OCs (e.g., PCBs, DDTs, and CHLs) that have a higher affinity for adsorption to particulates. With regard to BTs, as noted above, higher concentrations have been found in shallow-water fish than in deep-sea organisms (*36*). The butyltin profile shown in Figure 3b agreed with such a vertical trend reported in our previous study. Continuous input of BTs from the usage in surface waters (e.g., antifouling) would be well over the vertical transportation rate of these compounds, which results in higher levels in shallow waters.

In addition, the structure of the watermass around the western North Pacific and horizontal distribution of myctophids should be considered in evaluating the accumulation profiles. It has been suggested that the North Pacific Intermediate Water (NPIW), which is characterized by the salinity minimum and found at depths of 300−800 m in the North Pacific subtropical gyre, originates from the watermass in the Okhotsk Sea, and new NPIW is formed along the Kuroshio−Oyashio interfrontal zone (*58*, *59*). Elevated background levels of OCs in the Okhotsk Sea have been reported in several recent studies; higher concentrations of PCBs and DDTs were found in air and waters in East Siberia regions than those from the western North Pacific around Japan (*60*). Elevated concentrations of OCs in larga seals from the Okhotsk Sea (*61*) and sea eagles from Hokkaido, Japan (*62*), also suggested the high contamination status in this region.

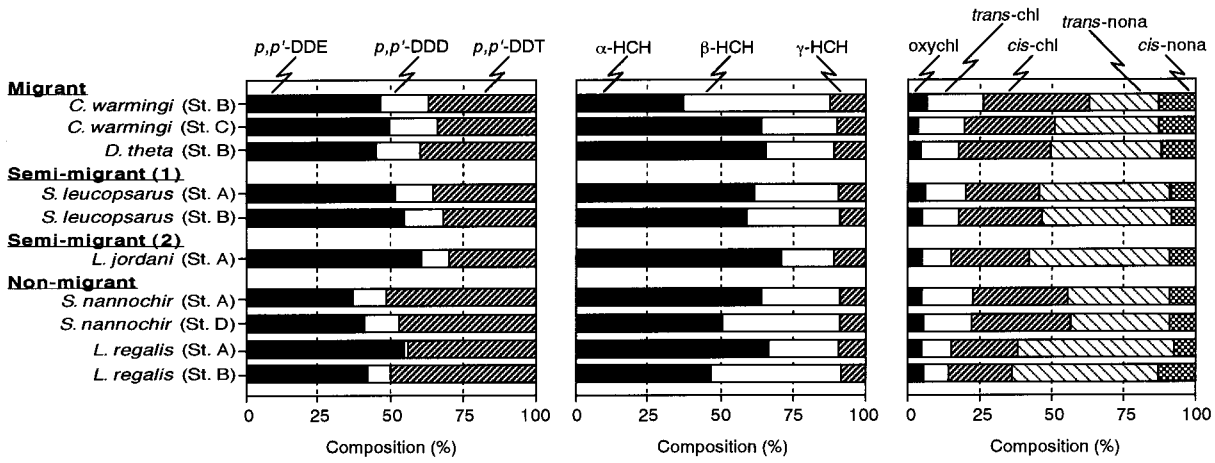

FIGURE 4. Compositions of DDT compounds, CHL compounds, and HCH isomers in myctophid fishes from the western North Pacific off northern Japan. Oxychl, *trans*-chl, *cis*-chl, *trans*-nona, and *cis*-nona indicate oxychlordane, *trans*-chlordane, *cis*-chlordane, *trans*-nonachlor, and *cis*-nonachlor, respectively.

The sampling depths (200−700 m) and locations (38−41° N, 143−147° E) for this study are almost identical with the area where a intrusion of watermass from the Okhotsk Sea into the mesopelagic zone and the formation of NPIW occurs (*58*, *59*). In this context, the intermediate water in the study area can be expected to have higher concentrations of OCs such as PCBs and DDTs that originate from more contaminated Okhotsk Sea. Interestingly, *C. warmingi*, which is distributed in the warmer Kuroshio waters and migrate to transitional waters between subarctic and subtropical waters (*39*), showed the lowest concentrations of PCBs, DDTs, and CHLs (Figure 3a). All other myctophids (e.g., *L. regalis, S. nannochir, D. thata, S. leucopsarus,* and *L. jordani*), which are distributed in subarctic and transitional waters along the Oyashio current (*39*), showed higher concentrations. Thus, *C. warmingi* may reflect less contaminated status by OCs in the warmer Kuroshio waters as compared to those taken along the Oyashio current.

The exceptionally higher concentrations of OCs and BTs found in *D. theta* can be explained by both the distribution (as discussed above) and the feeding behavior of this fish. Among the migrating species, *D. theta* actively feed in epi- and mesopelagic zones during day and night, respectively, and show the highest feeding rate (unpublished data). Thus, this species can be exposed to contaminants in both deep (PCBs, DDTs, and CHLs) and surface waters (HCHs, HCB, and BTs) and accumulate OCs and BTs at high concentrations. In contrast to *D. theta*, semi-migratory *S. leucopsarus* showing low concentrations of PCBs, DDTs, and CHLs feed only at night (epipelagic zone where low concentration of these compounds is expected), and its daytime feeding activity in mesopelagic zone is low. Thus, it seems reasonable that the most apparent difference was observed in the accumulation profiles between nonmigratory (*L. regalis* and *S. nannochir*) and semi-migratory fishes (*S. leucopsarus*); the former fish living in deep waters are not exposed to contaminants present in the surface waters, while the minimum exposure in deep waters is expected for the latter one because of their lower feeding activity in deep waters.

Composition of OCs and BTs. Among DDT compounds, *p,p′*-DDE or *p,p′*-DDT were predominat, and *trans*-nonachlor and α-HCH were the major constituents of chlordane compounds and HCH isomers, respectively, detected in myctophids (Table 2 and Figure 4). Among myctophids analyzed, higher proportions of *p,p′*-DDT were observed in nonmigratory fishes than in those of migratory ones ($p < 0.05$, Mann−Whitney *U*-test), while no significant difference among the migration types or sampling localities was found in the compositions of HCH isomers and chlordane compounds (Figure 4). This partly agrees with the concentration profile of DDTs as noted above but not with those of HCHs and CHLs, probably due to low concentrations of individual isomers and compounds.

Compared to the composition of DDT compounds found in deep-sea fishes from Suruga Bay (*24*), myctophids showed a higher proportion of *p,p′*-DDT and α-HCH ($p < 0.01$, Mann−Whitney *U*-test). This suggests the difference in contamination status and sources of DDTs and HCHs between Suruga Bay, a deep coastal body of water affected by the inflow of Kuroshio current (*64*), and the open waters of the western North Pacific near the Kuroshio−Oyashio transitional zone. The composition of chlordane compounds was not significantly different between myctophids and deep-sea fishes from Suruga Bay. Although the concentrations of BTs in myctophids were somewhat lower to discuss the composition in detail, TBT was the predominant compound in myctophids (Table 4). It may be attributable to a continuous input of TBT into the surface waters, which is likely from antifouling paints.

The mean percentage of *p,p′*-DDT (40 ± 7.6%) in myctophids collected in 1994 was apparently lower than those in myctophids (*D. suborbitalis*) collected from Yaizu, Suruga Bay, in 1976 (58%) (*47*). This suggests the reduction of fresh input of DDTs in the western North Pacific during last few decades. The decreasing *p,p′*-DDT proportion in total DDT compounds has also been reported in the study on temporal trend of OCs in northern fur seals from the Pacific Coast of Japan (*40*). In contrast to the composition of DDT compounds, no significant difference in the proportion of α-HCH was observed between the myctophids collected in 1976 (60%) and 1994 (58 ± 10%). As noted above, decreasing rate of HCHs during this period was smaller than those for DDTs. Such a small variation in the temporal trend of HCH isomer composition has also been reported in the northern fur seals (*40*). It may be attributable to the global transport of HCHs to high latitude regions and/or the input from local sources around the Okhotsk Sea (*60*−*62*, *64*). Further monitoring in the western North Pacific Ocean including marginal seas such as the Okhotsk Sea and Japan Sea along with geophysical and geochemical studies are required to delineate the distributions and fate of anthropogenic contaminants.

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