Contamination by Persistent Organic Pollutants and Related Compounds in Deep-Sea Ecosystems Along Frontal Zones Around Japan

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Abstract During the last few decades, contamination by anthropogenic chemicals such as persistent organic pollutants (POPs) has spread all over the world as evidenced by their detection in various environmental components and biota including those far from human activities. Particularly, research efforts on field observations and numerical models of global fate of POPs have revealed oceanic water bodies to be a global reservoir and final sink for these toxic contaminants that undergo transport from emission sources and partition between air and water and scavenge to deep-sea layers by various biogeochemical and geophysical processes. This chapter provides an overview of the contamination by POPs and related compounds in deep-sea ecosystems along frontal zones around Japan based on the results of the monitoring studies conducted by our laboratory during the last

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decade. In the chapter, we focus mainly on two regions, the western North Pacific (WNP), off-Tohoku, Japan, and the East China Sea (ECS). The WNP is a region influenced by various water masses and currents, making it one of the world's most biologically productive zones. The other region discussed in the chapter, the ECS, is an epicontinental sea with lots of continental inputs. Our studies in these regions were conducted with the objective of understanding the environmental transport and distribution and the specific accumulation characteristics of organohalogen and butyltin compounds in deep-sea organisms. Our results suggest the vertical transport of POPs and related compounds in high productive waters along oceanic fronts and the potential role of deep-sea bed as a final sink and reservoir for these persistent contaminants. Further, to implement and evaluate the effectiveness of international agreements to protect the marine environment from the deleterious effects of POPs, interdisciplinary approaches including studies on biogeochemical and geophysical processes in the ocean as well as field observations are required to delineate the global and regional fate of POPs.

Keywords Deep-sea ecosystem, East China Sea, Persistent organic pollutants, Western North Pacific

Contents

1   Introduction

Over the past few decades, large amounts of organic and inorganic contaminants have been released into the environment as a consequence of the worldwide urbanization and agricultural and industrial activities. Marine ecosystems are no exception and they have also been increasingly subject to anthropogenic chemical contamination, receiving contaminants from a variety of diffuse and point sources. The contaminants include industrial chemicals inadvertently released into the
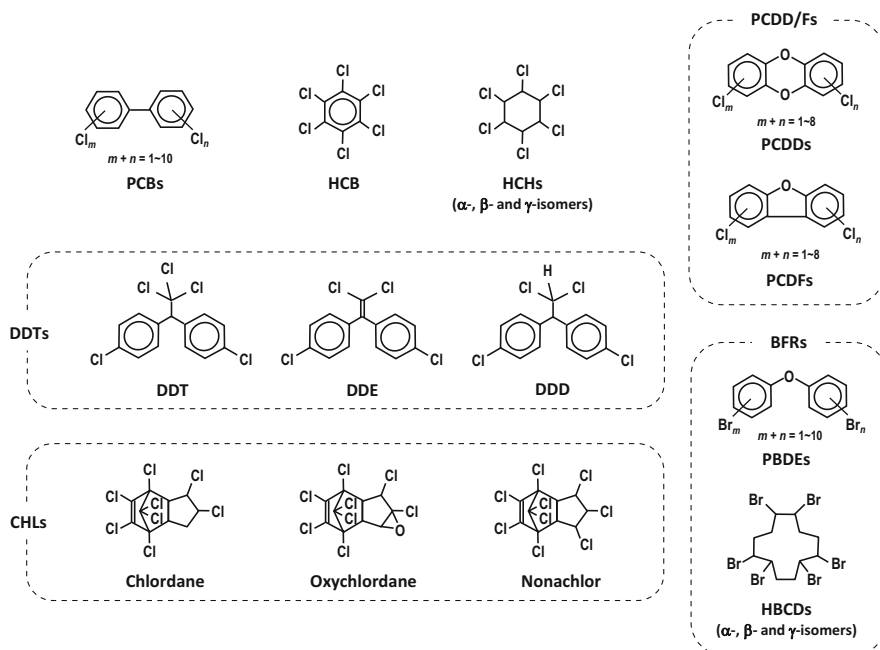
Fig. 1 Chemical structures of persistent organohalogen compounds (major POPs listed in the Stockholm Convention)

environment, as well as those derived from land use activities such as agricultural chemicals applied on crops. One such group of contaminants is the persistent organic pollutants (POPs), comprising various well-known organohalogen contaminants such as polychlorinated biphenyls (PCBs), polychlorinated dibenzo-*p*-dioxins and furans (PCDD/Fs), and organochlorine pesticides like dichlorodiphenyl-trichloroethane (DDT) and its stable metabolites, DDE and DDD (DDTs), chlordane-related compounds (CHLs), hexachlorobenzene (HCB), and hexachloro-cyclohexanes (HCHs) (Fig. 1). Besides, certain brominated organic compounds have been considered for inclusion in the list of POPs, i.e., polybrominated diphenyl ethers (PBDEs) and hexabromocyclododecanes (HBCDs) (Fig. 1). These compounds, which are added to electrical and electronic equipment, paints, textiles and building materials as brominated flame retardants (BFRs), are an emerging class of contaminants. In addition to the above contaminants, aquatic pollution by butyltins (BTs), particularly toxic tributyltin (TBT), used as a biocide in antifouling paints for boats and aquaculture nets, has been of concern due to the bioaccumulative potential and deleterious effects of BTs in organisms [1].

Both the emerging and legacy POPs are toxic, are chemically stable, and therefore do not easily degrade in the environment or in organisms. These chemicals tend to partition between various environmental media, such as air, water, soil, sediment, and biota, depending on the physicochemical properties.

Furthermore, being semi-volatile, these compounds are subject to co-distillation processes and can be transported globally through the atmosphere, which is well known as a concept of long-range atmospheric transport (LRAT) [2]. As a consequence, these substances have been detected in remote, relatively pristine locations, such as the polar regions and the deep seas. To protect human health and the environment, national and international control measures on the production and usage of POPs have been (or are being) introduced to reduce their emissions to the environment. For instance, the Stockholm Convention, negotiated under the support of the United Nations Environment Program (UNEP), established a globally binding framework initially targeting a group of POPs, informally called the "dirty dozen," for reduction and eventual elimination [3]. The dirty dozen include several organochlorine pesticides (aldrin, chlordane, dieldrin, endrin, heptachlor, hexachlorobenzene, mirex, toxaphene, and DDT), PCBs, and PCDD/Fs. Besides the pesticide use, HCB was also used for industrial processes. PCBs were widely applied for industrial fluids in electrical equipment. PCDD/Fs and some PCB congeners (i.e., dioxin-like PCBs or coplanar PCBs) are formed as unintentional by-products during incineration and other thermal processes. The Stockholm Convention also includes procedures for identifying and adding substances to the POPs list. In May 2009, nine other POPs, including several BDE congeners present in PBDE commercial mixtures, have been listed under the Stockholm Convention [4]. More recently, HBCD has also been listed to Annex A of the Stockholm Convention at the sixth Conference of Parties (COP 6) in 2013 [5].

Despite a limited number of studies, significant contamination by POPs in deep-sea fishes has been demonstrated in the early 1980s [6–9]. Besides, in recent years, POPs, including BFRs, have been found at appreciable concentrations in deep-sea organisms from various parts of the world [10–28]. Research efforts by various scientific groups in the last few decades have greatly increased our understanding on the global distribution of POPs. The world's oceans are thought to play an important role in the cycling and removal of POPs [29]. The oceans cover two-thirds of the Earth's surface and because of their large volume can contain a large inventory of POPs [30]. The sources of POPs to the marine environment are riverine transport, municipal and industrial discharges, continental runoff, and atmospheric deposition in open waters. Atmospheric deposition delivers a large proportion of the POPs present in the oceans, through various mechanisms such as diffusive air–water exchange of POPs and wet and dry deposition processes [31]. Once in the aquatic environment, POPs can be dissolved in the water phase or partition onto colloidal and suspended/settling particulate matter, incorporate into food webs, transfer to the deep waters with the sinking particles, and eventually deposit in bottom sediments. It should be noted as an important role in the global behavior of POPs that the bottom sediments of deep-sea can either act as a final sink or a reservoir for such persistent contaminants. Sediment-sorbed xenobiotics can be taken up by the epibenthic and infaunal biota as they feed. A limited desorption of POPs, independent from that mediated by the bottom currents at the superficial level of the sediment, also occurs in the interstitial water [32]. These features can lead to remobilization of sediment-bound POPs in the entire food web of the
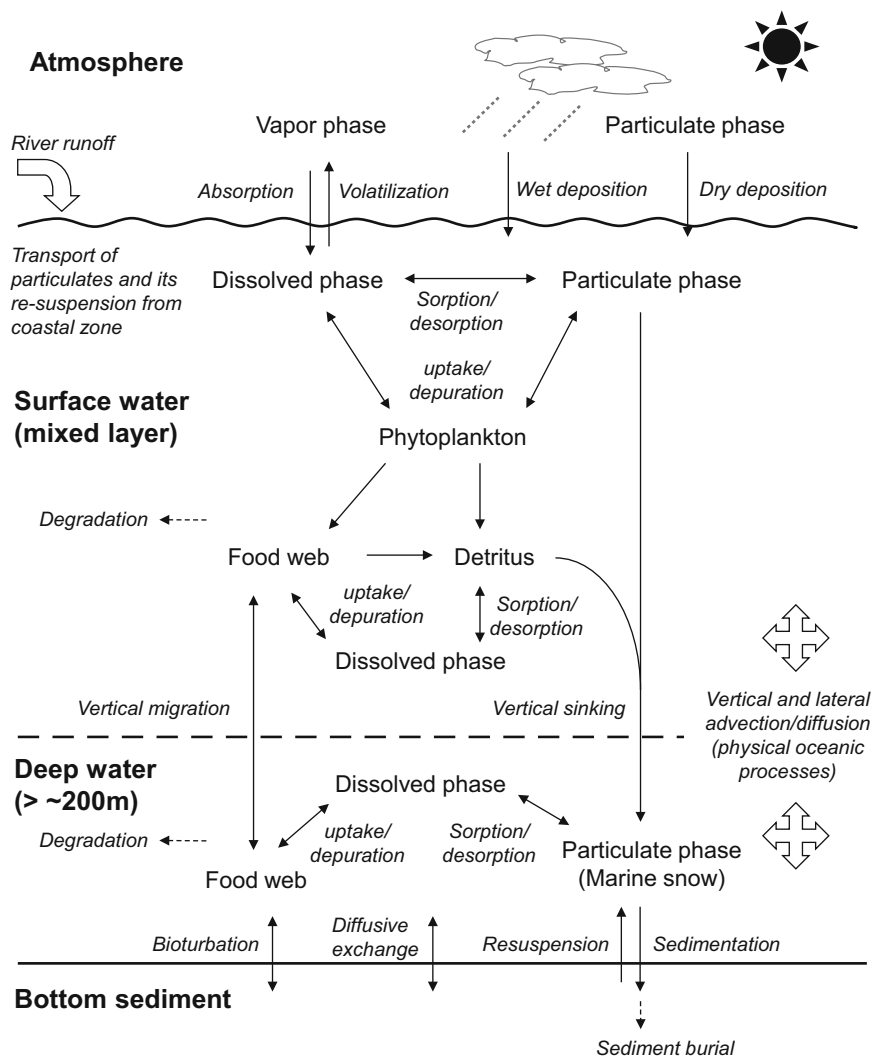
Atmosphere

Vapor phase   Particulate phase

River runoff

Absorption │ *Volatilization*   *Wet deposition*   *Dry deposition*

*Transport of
particulates and its
re-suspension from
coastal zone*   Dissolved phase   ← *Sorption/* →   Particulate phase
                                          desorption

                                          uptake/
                                          depuration

**Surface water
(mixed layer)**

                        Phytoplankton

Degradation ←----

                Food web   →   Detritus

                    uptake/   *Sorption/*
                    depuration   *desorption*

                        Dissolved phase

Vertical migration   *Vertical sinking*   *Vertical and lateral
                                                                   advection/diffusion
                                                                   (physical oceanic
                                                                   processes)*

**Deep water
(> ~200m)**
                        Dissolved phase

Degradation ←----   *uptake/*   *Sorption/*
                          depuration *desorption*   Particulate phase
                Food web   (Marine snow)

                        Diffusive
Bioturbation   *exchange*   *Resuspension*   *Sedimentation*

Bottom sediment

                                        Sediment burial

Fig. 2 Conceptual diagram showing the key processes affecting the transport of POPs between the atmosphere, water column, and bottom sediments

deep-sea benthos. Further, the fate of POPs are affected by the hydrodynamics of water masses such as turbulence and advection of water masses and the differential characteristics of coasts with respect to the open sea, i.e., enhanced stratification due to freshwater input from rivers, influence of tides etc. Figure 2 is a conceptual diagram showing the key processes affecting the transport of POPs between the atmosphere, water column, and bottom sediments. Depending on the physico-chemical properties of POPs (e.g., log K_{aw}), their global partition between

environmental media and transport processes differ significantly among the compounds, congeners, or isomers [31, 33].

For the past four decades, our laboratory at the Center for Marine Environmental Studies (CMES), Ehime University, Japan, has been conducting research on the contamination status and spatial distribution, temporal trends, behavior, and fate of the above-listed contaminants in various matrices from different ecosystems (reviewed in Tanabe [34]; Tanabe et al. [35]; Tanabe and Ramu [36]). In the following chapter, we review some of our studies dealing with POPs in deep-sea organisms collected from various deep-sea environments. Although it is ideal to directly measure the levels of POPs in seawater to monitor the contamination status of POPs in marine ecosystems, there are limitations such as large volumes of seawater required for the analysis, sensitive analytical techniques crucial for detecting the low concentrations of POPs, and fluctuations of POP concentrations in seawater depending on the weather conditions. Therefore, in our studies, we often employ marine organisms like fish and shellfish as bioindicators to elucidate contamination status and spatial distribution of POPs in marine ecosystems. Aquatic organisms are very efficient in accumulating these contaminants, since, in addition to bioaccumulation through the diet, they are also subject to bioconcentration. Furthermore, data regarding levels and distributions of POPs in marine organisms, especially edible ones (and this includes an increasing number of deep-sea species), are important not only for assessing the state of the ecological environment but also from the human health perspective.

In the chapter, we focus mainly on two regions, the western North Pacific (WNP), off-Tohoku, Japan, and the East China Sea (ECS). The WNP is a region influenced by various water masses and currents making it one of the world's most biologically productive zones. The other region discussed in the chapter, the ECS, is an epicontinental sea with lots of continental inputs. Both the regions are characterized by the formation of numerous oceanic fronts. Belkin [37] defined oceanic fronts as a narrow zone of enhanced horizontal gradients of water properties (temperature, salinity, nutrients, etc.) that separates broader areas with different water masses or different vertical structure. These fronts are mostly characterized by strong mixing, stirring, enhanced bioproductivity, and ecotones. High productivities around the fronts may enhance flux of POPs into the oceans and their transport to deep waters because of phytoplankton uptake and the vertical flux of the particles play important roles in the biogeochemical cycles of POPs in the oceans [38, 39]. In general, land- and open-ocean-derived materials tend to converge at the frontal zones. The formation of coastal fronts has been suggested to be an important oceanic phenomenon determining the behavior and flux as well as the fate of persistent contaminants in the marine environment [40].

Detailed information on the sampling and analytical methods can be found in the cited papers [14, 41–44]. The reviewed studies may help improve our insights into the contamination status and the fate of legacy POPs and emerging contaminants such as BFRs in the deep-sea environments.

Fig. 3 A schematic circulation pattern of the major currents and frontal structures in the Kuroshio–Oyashio transition area, *EKC* East Kamchatka Current, *ESC* East Sakhalin Current, *OY* Oyashio, *TWC* Tsushima Warm Current, *KE* Kuroshio Extension, *OYF* Oyashio Front, *PF* Polar Front, *SAF* Subarctic Front, *OSMW* Okhotsk Sea Mode Water
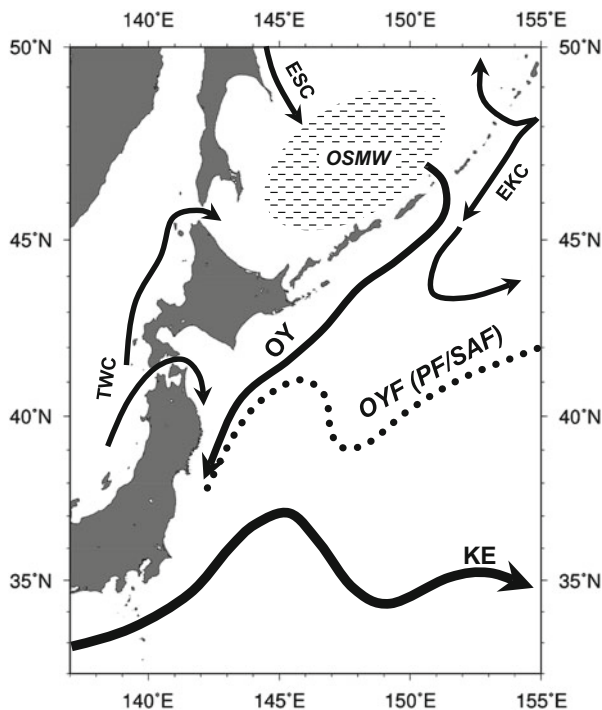
2 Description of the Study Areas

2.1 The Western North Pacific, Off-Tohoku

The study area, the WNP, off-Tohoku, is characterized by a complex oceanographic structure represented by major ocean currents (Fig. 3). The Oyashio, a western boundary current of the Subarctic North Pacific, is a continuation of the East Kamchatka Current (EKC) and is fed by waters from the Western Subarctic Gyre and the Sea of Okhotsk [45]. The primary productivity of the Sea of Okhotsk is very high, especially on the continental shelf due to the relatively high insolation and the nutrient input from the Amur River and Pacific Ocean [46]. Recently, the dense shelf water flowing into Okhotsk Sea Mode Water (OSMW) has been considered as a potential source for exporting large amounts of organic matter and nutrients such as iron from the continental shelf to adjacent ocean interior along Oyashio [47, 48]. The eastward flowing Oyashio forms the Oyashio Front (OYF) which becomes Polar Front (PF) or Subarctic Front (SAF) to the east. The Kuroshio, a western boundary current from the southern tropic area, turns eastward from the eastern coast of Honshu, Japan. Then, warm and saline water is transported by this Kuroshio Extension [49]. The region between the Oyashio Front and the Kuroshio Extension Front is called the Kuroshio–Oyashio Transition Zone (KOTZ) or mixed water region, in which cold and warm waters mix and complex frontal structures are formed [49, 50].

Thus, the oceanography in the Tohoku area is complex and variable due to the confluence of various currents and water masses. Due to mixing of these water masses, the nutrient-rich subsurface water is brought into the euphotic zone promoting high phytoplankton production [51]. Marine biogeochemical activities including primary production, zooplankton grazing, microbial transformation, aggregation, and degradation processes of organic particles influence the flux of biogenic particles in the ocean. Vertical sinking of particle-associated pollutants such as PCBs may be enhanced by eutrophication since higher primary productivity leads to larger vertical fluxes of particles and organic carbon [38, 52]. This region known as Japan Trench is a depository of biogenic and lithogenic materials that are transported from the coastal shelf [53]. Thus, it can be assumed that a considerable proportion of anthropogenic contaminants may be transported into the deep water of North Pacific through the various biogeochemical processes.

2.2   The East China Sea

The ECS, located at midlatitudes between 25 and 35°N is an epicontinental sea surrounded by the Ryukyu archipelago, Japan, Korea, China, and Taiwan (Fig. 4). It has a broad continental shelf covering an area of 530×10^3 km² [54]. The major western boundary current, the Kuroshio, runs along the outer edge of the continental shelf, enters the ECS through the strait between Taiwan and the westernmost island of the Ryukyu Islands, flows northeastward along the shelf slope, and exits to the Philippine Sea after turning eastward near 30°N [55]. Two of the largest rivers in the world, the Yangtze River (Changjiang) and the Yellow River, discharge into the ECS. Thus, the cold, freshwater distributed on the continental shelf and the warm saline Kuroshio water that occupies the area around the shelf water lead to the formation of salinity front near the continental shelf break.

The Kuroshio, the Yangtze River runoff, and the East Asia monsoons are the dominant factors affecting the circulation in the ECS. The ECS continental shelf circulation pattern is characterized by the Kuroshio, Tsushima Current, Taiwan Warm Current, and other coastal waters and shelf fronts [56, 57] (Fig. 4). It has been demonstrated that the Kuroshio strongly influences not only the circulation in the ECS shelf but also its chemistry through water-mass exchanges [56–58].

Riverine runoff is an important mode to transport anthropogenic pollutants from terrestrial sources to adjacent oceans. The Yangtze River flows through densely populated areas with agriculture and industrial activities along both the banks, and Shanghai, the largest city in China, is situated at its mouth. Discharge of industrial wastes, application of fertilizers, pesticides, and herbicides in farming, as well as heavy metal pollution are said to make the Yangtze River one of the most polluted rivers in the world [59]. Li and Daler [60] reported that the environmental pollution of the Yangtze River basin greatly influences the state of the marine environment of the ECS. Consequently, the ECS has attracted much interest as a site for the study of the fate of terrestrial material in the marine environment [61]. The ECS shelf receives a rich supply of nutrients from the Yangtze River and the upwelled Kuroshio subsurface waters. As a result, the ECS shelf is one of the most productive

Fig. 4 A schematic circulation pattern of the major currents and frontal structures in the East China Sea. *TsC* Tsushima Current, *TWC* Taiwan Warm Current, *WKCC* Western Korea Cold Current, *CDW* Changjiang Diluted Water, *ECSCoW* East China Sea Coastal Water, *YSCoW* Yellow Sea Coastal Water, *YSWW* Yellow Sea Warm Water. *Dashed lines* indicate major shelf fronts in the East China Sea
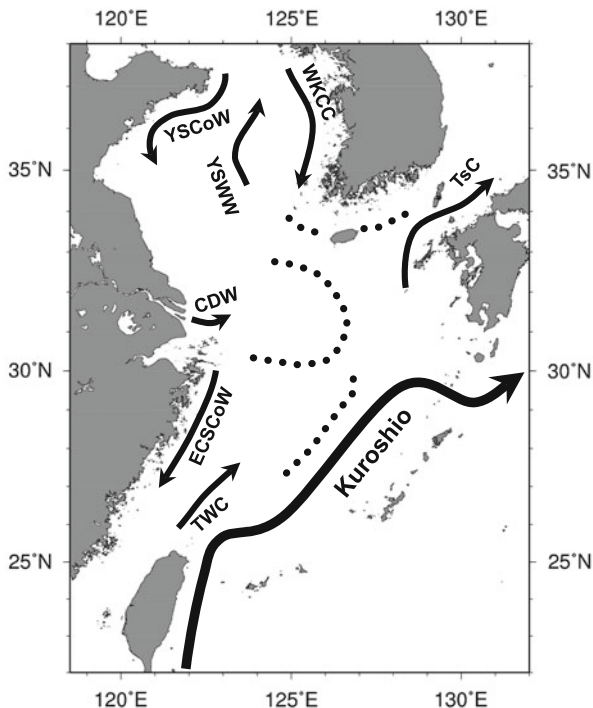
marginal seas in the world [62]. Thus, it is obvious that the ECS receives enormous amounts of anthropogenic pollutants, suspended matter, and nutrients with the riverine runoff. Furthermore, the intensive exchange between the shelf water and the Kuroshio and the high primary productivity observed in this region may facilitate the flux of persistent contaminants to the bottom of the ECS.

3 POPs and Related Compounds in the Western North Pacific, Off-Tohoku

This section provides a synopsis of our studies conducted in the WNP, off-Tohoku [14, 41, 44], with the objective of understanding the distribution and the specific accumulation characteristics of organohalogen and butyltin compounds in deep-sea organisms in this region.

3.1 Contamination Status

Organochlorine compounds (OCs) were detected in all the deep-sea organisms (e.g., deep-sea eels, grenadiers, cods, eelpouts, sculpins, bikumins, flounders, myctophids,

lantern sharks, ratfishes, squids, octopus, shrimps, crabs, gastropods) collected during three different time periods (see the above references for more details). The concentrations of PCBs and DDTs were the highest among the OCs analyzed, and the concentrations of other compounds were in the following order CHLs > HCHs ≥ HCB. The predominant accumulation of PCBs and DDTs in deep-sea organisms collected from the WNP, off-Tohoku, agreed with those reported in shallow- and deep-water fishes collected from other locations along the Pacific coast of Japan [12, 15] (Table 1). This reflects the higher bioaccumulative properties of PCBs and DDTs in marine food web as well as their significant usage.

Except for some higher trophic level organisms like snubnosed eels (*Simenchelys parasitica*), which contained some OCs at extremely high concentrations, the concentrations of OCs in deep-sea fish collected from the WNP, off-Tohoku, were generally lower than those in deep-sea organisms collected from other locations in the Atlantic Ocean, Mediterranean Sea, Ireland and Norwegian coasts, and the Arctic region [10, 11, 17–21, 25–27] (Table 2). The relatively low contamination by OCs in the present study area might be due to the smaller usage of these compounds in the WNP region compared to that of the North American and European countries. Besides, factors like variations in analytical methods and fish species (having different biological and ecological characters) may also influence the differences in the OC levels compared between the studies.

Comparing with studies carried out in adjoining areas on deep-sea fishes by our research group (Table 1), the concentrations of PCBs, DDTs, CHLs, and HCB in deep-sea fishes collected from the WNP, off-Tohoku, were comparable or lower than the data so far reported from Suruga and Tosa Bays [12, 15] and the ECS [42]. On the other hand, concentrations of HCHs in deep-sea fishes from this region were higher than those from other locations along the warm Kuroshio Current such as Tosa Bay and the ECS (Table 1). Higher concentrations of HCHs in cold waters along the Oyashio Current than in other offshore waters around Japan were also observed in a study using skipjack tuna (*Katsuwonus pelamis*) [63]. Due to the high vapor pressure, HCHs are known to rapidly evaporate and be transported from their pollution sources in the tropics and temperate regions to colder regions via the atmosphere [2, 30, 64]. The distribution patterns of HCHs found in the deep-sea organisms reflect the highly transportable nature of HCHs and its accumulation in the cold-water current of the WNP. No significant difference in the contamination status of HCHs between three research periods from 1994 to 2005 in this region (Table 1) is also suggestive of continuous flux of HCHs into the cold waters of the WNP region.

Among the three studies reviewed here, only the study by Takahashi et al. [44] reported the concentrations of PBDEs and HBCDs in deep-sea fishes collected from the western North Pacific, off-Tohoku, in 2005. The concentrations of PBDEs ranged from 1.3 to 8.5 ng/g with a mean of 3.6 ng/g lipid wt, while the concentrations of HBCDs ranged from 5.4 to 45 ng/g with a mean of 22 ng/g lipid wt (except for snubnosed eels). Despite the low levels compared to the other POPs, the detection of PBDEs and HBCDs in deep-sea fishes indicates the widespread presence of such "emerging POPs" even in deep oceans and their long-range transport. Similar to the

Table 1 Comparison of mean and range concentrations of organochlorines (ng/g lipid wt) between fishes from the western North Pacific and East China Sea

Location	Sampling year	PCBs	DDTs	CHLs	HCHs	HCB	References
Off-Tohoku myctophids	1994	160 (20–370)	110 (15–280)	22 (5–47)	13 (3.1–24)	4.0 (0.79–8.0)	[14]
Off-Tohoku deep-sea fish[a]	1995	420 (n.d. to 2,200)	230 (14–830)	110 (3.9–640)	30 (n.d. to 150)	16 (n.d. to 100)	[41]
Off-Tohoku deep-sea fish[a]	2005	150 (34–390)	110 (36–220)	54 (19–120)	25 (5–69)	21 (7–45)	[44]
Tosa Bay shallow-water fish	1997	310 (n.d. to 1,100)	220 (38–1,200)	46 (n.d. to 200)	5.3 (n.d. to 14)	5.7 (n.d. to 13)	[15]
Tosa Bay deep-sea fish	1997	340 (n.d. to 1,600)	290 (7.1–1,200)	44 (5.6–220)	11 (n.d. to 21)	11 (n.d. to 61)	[15]
Suruga Bay shallow-water fish	1993–1994	1,600 (540–2,600)	390 (80–1,700)	140 (46–320)	4.1 (n.d. to 8.0)	8.8 (3.7–24)	[12]
Suruga Bay deep-sea fish	1993–1994	1,000 (450–1,900)	390 (51–910)	260 (69–770)	25 (10–37)	20 (9.9–50)	[12]
East China Sea shallow-water fish	2001–2003	150 (20–830)	330 (110–1,200)	39 (5.9–180)	11 (2.2–62)	17 (n.d. to 70)	[42]
East China Sea deep-sea fish	2001–2003	230 (36–1,400)	720 (n.d. to 7,900)	40 (3.7–240)	11 (1.4–41)	18 (1.7–290)	[42]

Figures in parentheses indicate the range of concentrations

n.d. not detected (for calculation of mean, data less than detection limits was assumed to be 0)

[a]Data without snubnosed eel

Table 2 Comparison of mean concentrations of PCBs and DDTs in deep-sea fishes from various parts of the world

Species	Sampling year	Location	Depth (m)	PCBs (ng/g) wet wt	PCBs (ng/g) lipid wt	DDTs (ng/g) wet wt	DDTs (ng/g) lipid wt	References
Cartilaginous fish (2 sp.)	1992	Davis Strait (Greenland)	800–2,200	310	430	490	690	[10]
Bony fish (6 sp.)	1992	Davis Strait (Greenland)	200–2,100	300	790	250	670	[10]
Fish (6 sp.)	1993–1994	Suruga Bay, Japan	200–740	160	1,000	60	390	[12]
Cartilaginous fish (1sp.)	1992	Norway (Nordfjord)	400	1,800	2,400	4,600	6,000	[11]
Bony fish (3 sp.)	1992	Norway (Nordfjord)	400	3,900	8,700	8,800	20,000	[17, 18]
Armed grenadier	1992	North Atlantic	2,900	–	2,100	–	1,090	[17, 18]
Black halibut	1994–1998	South Atlantic	–	–	1,000	–	390	[17, 18]
Kingklip	1994–1998	South Atlantic	–	–	1,400	–	190	[17, 18]
Dover sole	1995	Monterey Bay Canyon	–	–	2,200	–	2,400	[17, 18]
Longspine thornyhead	1995	Monterey Bay Canyon	–	–	3,700	–	2,400	[17, 18]
Snubnosed eel	1995	Off-Tohoku, Japan	1,000	1,100	6,700	2,100	13,000	[41]
Fish (21 sp.)	1995	Off-Tohoku, Japan	150–1,300	81	400	30	220	[41]
Gadiform (*Mora moro*)	1996	Gulf of Lions	986–1,136	3,400	9,900	1,200	4,300	[19]
Fish (20 sp.)	1997–1998	Tosa Bay, Japan	150–400	39	350	31	290	[15]
Roundnose grenadier	1999	West of Ireland	1,000	450	770	730	1,300	[20]
Roundnose grenadier	1999	West of Ireland	2,000	800	1,900	1,000	2,400	[20]
Skate (3 sp.)	2000	Southern Adriatic Sea	–	310	890	–	–	[21]
Ghost shark	2000	Southern Adriatic Sea	–	280	390	–	–	[21]
Fish (37 sp.)	2001–2003	East China Sea	89–512	6.2	230	30	700	[42]
Fish (10 sp.)	2002	Sulu Sea	292–1,015	0.59	58	1.8	150	[22]
Hollowsnout grenadier	2003	Adriatic Sea	–	–	430	–	340	[25]

Roughtip grenadier	2003	Adriatic Sea	–	–	310	–	5,400	[25]
Hollowsnout grenadier	2006	Southern Adriatic Sea	–	–	1,200	–	760	[26]
Roughsnout grenadier	2006	Southern Adriatic Sea	–	–	12,000	–	5,400	[26]
Roundnose grenadier	2000–2002	Porcupine Seabight	1,000–1,900	–	2,400[a]	–	1,500[a]	[27]
Snubnosed eel	2005	Off-Tohoku, Japan	900	720	6,300	1,300	12,000	[44]
Fish (11 sp.)		Off-Tohoku, Japan	400–1,000	12	150	8.2	100	[44]

–: not available

[a]Median concentration

For calculation of mean, data less than detection limits was assumed to be 0

For comparison, all the data were rounded into two significant digits

result above, higher concentrations of HBCDs than PBDEs were observed in skipjack tuna collected from offshore waters around northern Japan [65, 66]. On the other hand, accumulation of PBDEs at higher concentrations than HBCDs was reported in fishes and cetaceans from East and South China Seas [35, 43]. Comparing to the concentration levels of PBDEs (5.1 and 19.9 ng/g lipid wt at mean concentrations) and HBCDs (<1.5 ng/g lipid wt) in two species of deep-sea grenadiers from the Mediterranean Sea [23], relatively high magnitude of contamination by HBCDs was noted in our data on deep-sea fish from the western North Pacific, off-Tohoku. Such differences among the studies and locations may reflect possible long-range transport of HBCDs to cold offshore waters, like HCHs, and/or significant usage of HBCDs (especially for thermal insulating materials) in northern Japan. Although HBCD is currently being proposed to be listed a POP in the Stockholm Convention, the contamination by this BFR in marine ecosystems around Japan may increase or continue in the meantime as its use or disposal, considering the increase in market demand for HBCD in Japan during late 1990s [67].

Concentrations of butyltin compounds (BTs) were reported by Takahashi et al. [14] and de Brito et al. [41]. Takahashi et al. [14] detected BTs in almost all myctophids analyzed (levels up to 46 ng/g wet wt for total BTs ($\sum$BT: MBT + DBT + TBT)). Widespread contamination by BTs in deep-sea organisms from the western North Pacific, off-Tohoku (levels up to 530 ng/g wet wt in the liver of demon grenadier, *Coelorinchus gilberti*, for $\sum$BTs), was also reported by de Brito et al. [41]. Although the magnitude of contamination by BTs in open-water ecosystems of the western North Pacific seems to be relatively low, the results indicate the expansion of BT pollution in deep-sea ecosystems.

3.2 Species-Specific Accumulation and Trophic Magnification

Among various fish species analyzed, snubnosed eel (Fig. 5) accumulated hydrophobic organohalogens (with log K_{ow} ~ 6–8) such as PCBs, DDTs, CHLs, and PBDEs at extremely high concentrations (Table 2). This species can be considered as an organism at a high trophic level in the food web due to its strong carnivorous and unique feeding habit; this fish has been found to burrow into the body of large fish such as halibut and shark and feed on their intestine and blood [68]. Relatively higher concentrations of these compounds were also found in some large carnivore and/or bottom-dwelling fishes. These observations suggest that accumulation of the hydrophobic OC and PBDEs is influenced by trophic magnification and/or feeding habits of organisms and their long biological half-lives, i.e., higher uptake rate via food and slower equilibrium between ambient water and body lipids. In contrast, HCHs, HCB, and HBCDs, which have less lipophilicity than the other organohalogen compounds, showed smaller variation in concentrations among species of different trophic levels and feeding habits.
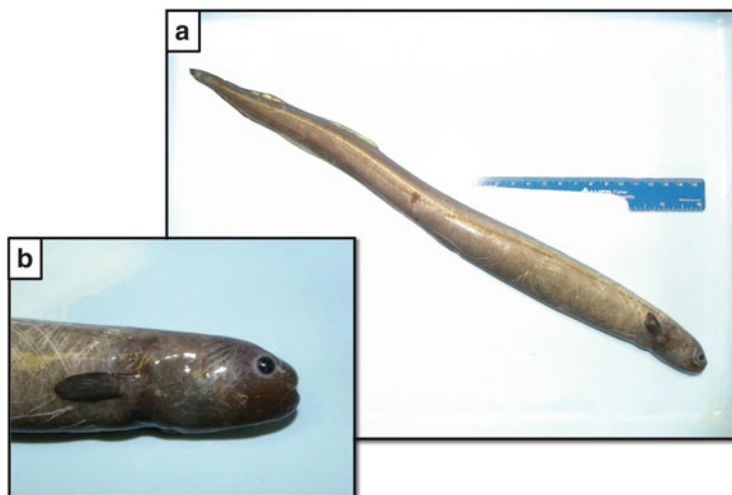
Fig. 5 Snubnosed eel (*Simenchelys parasitica*) (Photo by Dr. T. Kubodera, National Museum of Nature and Science, Tokyo, with permission). (**a**) Whole body; (**b**) head part

Isotopic signatures based on the ratios of different stable isotopes of elements have been broadly applied to investigate a variety of processes. In particular, stable carbon and nitrogen isotope ratios (δ^{13}C and δ^{15}N) have been used in environmental studies for delineating carbon flows and trophic relationships in continental and marine ecosystems. The stable isotope ratios of nitrogen δ^{15}N reflect diets and represents time-integrated averages of assimilated foods [69]; thus, stable nitrogen isotope analysis has been used to assess the relative trophic level of aquatic biota and provide a quantitative continuous variable for studying the trophic magnification of contaminants within complex food webs [70, 71]. In contrast, the ratios of stable carbon isotopes δ^{13}C, in biota, can help to elucidate trophic interactions by establishing the relative contribution of marine (or pelagic) versus coastal (or benthic) carbon sources [72]. Thus, the quantification of both stable carbon and nitrogen isotope ratios can provide valuable information on the feeding ecology of biota and its potential influence on the trophic enrichment of contaminants [70, 73].

To understand the trophic magnification of organohalogen compounds, δ^{15}N and δ^{13}C were determined in fishes collected in 2005. Their correlations to the concentrations of organohalogen compounds were discussed [44]. A wide range of δ^{13}C values among fishes was observed, implying the existence of multiple carbon sources in the food web of this region. Significant positive correlations were found between δ^{15}N (‰) and lipid normalized concentrations of PCBs, DDTs, and PBDEs ($p < 0.05$) showing their high trophic magnification potential in marine food webs (Fig. 6). No such relationship was found for HCB, HCHs, and HBCDs (Fig. 6). The rapid elimination through gills to surrounding water and by excretion via other means may prevent the trophic magnification of these compounds. In addition, less trophic magnification observed for some organohalogen
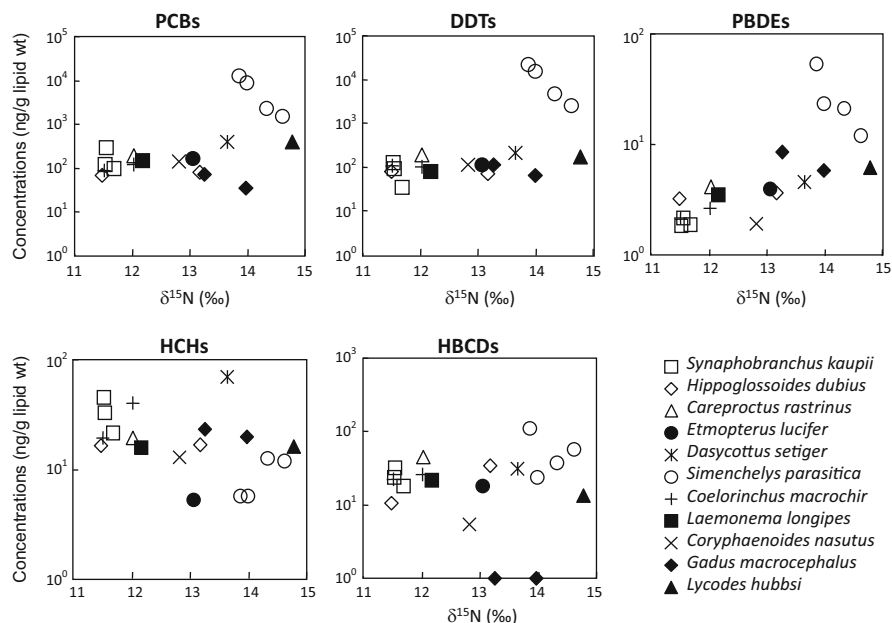
Fig. 6 Relationship between $\delta^{15}N$ (‰) and organohalogen compounds in deep-sea fishes from the western North Pacific, off-Tohoku, Japan

compounds in this study may be due to the smaller difference in trophic levels of fishes. In fact, there were no significant correlations between $\delta^{15}N$ values and the concentrations of almost all the compounds analyzed when the data of snubnosed eels was excluded (only a significant relationship for PBDEs with $p < 0.05$). Thus, the accumulation profiles of organohalogen compounds in deep-sea fishes reflect not only their trophic transfer but also the different sources of the contaminants in the food web and/or species-specific distribution of fishes. Especially, some bottom-dwelling fishes including snubnosed eels showed significant lower $\delta^{13}C$ values than the other fish species, reflecting influence of substances originated from sediments. Therefore, the data of high trophic bottom-dwelling fishes were excluded for further discussion on spatial and vertical distribution of contaminants.

A considerable variation was observed in the concentrations of BTs among tissues/organs and species. Among the tissues/organs analyzed, higher concentrations of BTs were found in the liver of fish (97 ± 212 ng $\sum$BT/g wet wt) and viscera of crabs (17 ± 17 ng $\sum$BT/g wet wt) [41]. Nevertheless, in the case of BTs, relatively high concentrations were observed not only in larger carnivore fishes (e.g., 9.7 ng $\sum$BT/g wet wt in the whole body of Pacific grenadier) but also in small fishes (up to 46 ng $\sum$BT/g wet wt in the whole body of myctophid fishes) and squids (up to 44 ng $\sum$BT/g wet wt in the whole body of Japanese hooked squid) [14, 41]. It is evident from previous studies that the bioaccumulation properties of BTs (i.e., organometallic compounds) were principally different from organohalogen

compounds having hydrophobic and/or lipophilic nature. Less biomagnification of BTs was also documented in some food webs along Japanese coasts [74, 75].

3.3   Distribution in Relation to the Water-Mass Structure

Even among fishes showing similar trophic levels, considerable variation in the concentrations of POPs and related compounds was observed, which cannot be explained only by their feeding habits and trophic magnification. It has been documented that fishes can be used as suitable bioindicators to elucidate the environmental levels and distributions of persistent and lipophilic compounds in water bodies after careful consideration of their biological and ecological parameters [76].

Takahashi et al. [14] studied the accumulation profiles of OCs and BTs in relation to the sampling locations and diel vertical migration modes of myctophid species to delineate the spatial and vertical distribution of the contaminants in the WNP. While concentrations of OCs and BTs varied among species, no significant difference in the concentrations of fishes were observed between sampling locations (in the area: 38–41°N, 143–147°E). On the other hand, the accumulation patterns of contaminants showed a specific trend in accordance with the migration types (Fig. 7). Except for a migrant species *D. theta*, higher concentrations of PCBs, DDTs, and CHLs were found in non- or less-migratory species living in deeper waters. On the contrary, HCHs, HCB, and BTs accumulated at higher concentrations in migratory species as compared to nonmigratory fishes. A similar trend can also be suggested from mean concentrations of OCs and BTs in deep-sea fishes collected from the WNP, off-Tohoku [41] (Fig. 8). In general (except for snubnosed eels), higher concentrations of PCBs, DDTs, and CHLs were found in fishes from deeper waters (~1,000 m). On the contrary, HCHs, HCB, and BTs accumulated at higher concentrations in fishes from shallower waters.

These observations suggest the vertical distribution of these contaminants in the water column of this region. Tanabe and Tatsukawa [77] 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. Long-range atmospheric transport and cold condensation of volatile POPs, as noted in the above section, may enhance their flux to the surface waters in this WNP region. On the other hand, results of these studies are also suggestive of the vertical transport of hydrophobic OCs and their accumulation in benthic deep-sea organisms. OCs with lower water solubility and higher affinity for adsorption to particles would be transported to deeper waters and incorporated into the marine food web more readily than more water-soluble compounds. With regard to BTs, higher concentrations were found in shallow-water than deep-sea fishes. Recent input of BTs from the usage in surface waters (e.g., antifouling) could be well over the vertical transportation rate of these compounds, which results in higher levels in shallow waters.

In addition, the structure of the water mass around the WNP should be considered in evaluating the vertical profiles of contaminants. As noted in the description of the studied area, it has been suggested that the North Pacific Intermediate Water
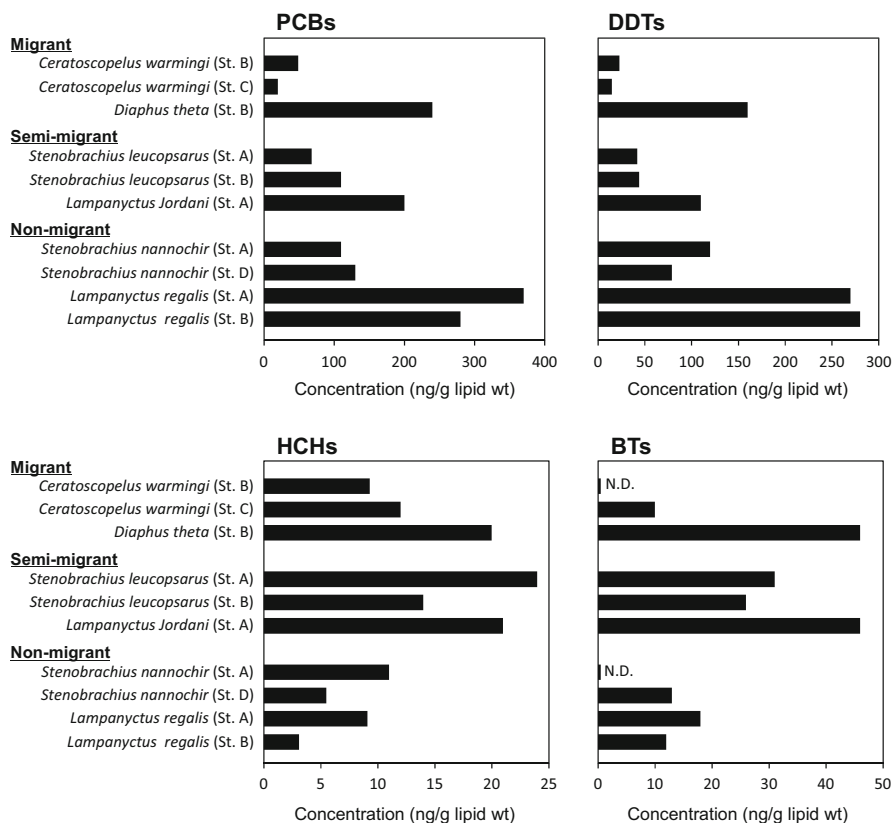
Fig. 7 Concentrations and residue patterns of PCBs, DDTs, HCHs, and BTs in myctophid fishes from the western North Pacific, off-Tohoku, with different migration types. Each *bar* indicates the concentration in a composite sample which was prepared from pooled and homogenized individual specimen belonging to the same species collected from the same sampling location [14]

(NPIW), which is found at depths of 300–800 m in the North Pacific subtropical gyre, originates from the water mass of OSMW, and new NPIW is formed along the Kuroshio–Oyashio interfrontal zone [50]. The dense shelf water flowing into the OSMW has been considered as a potential source for exporting large amounts of organic matter and nutrients such as iron from the continental shelf to adjacent ocean interior along Oyashio. This means that organic pollutants as well as the shelf-originated nutrients can be transported from the continental shelf to NPIW. Despite limited data available, elevated background levels of OCs such as DDTs and PCBs in the Okhotsk Sea and East Siberia regions have been suggested by several studies on air and water [78, 79] and wild animals such as seals and sea eagles [80, 81]. The sampling depths and locations in the study of Takahashi et al. [14] and de Brito et al. [41] overlapped with the area where an intrusion of water mass from the Okhotsk Sea into the mesopelagic zone and the formation of
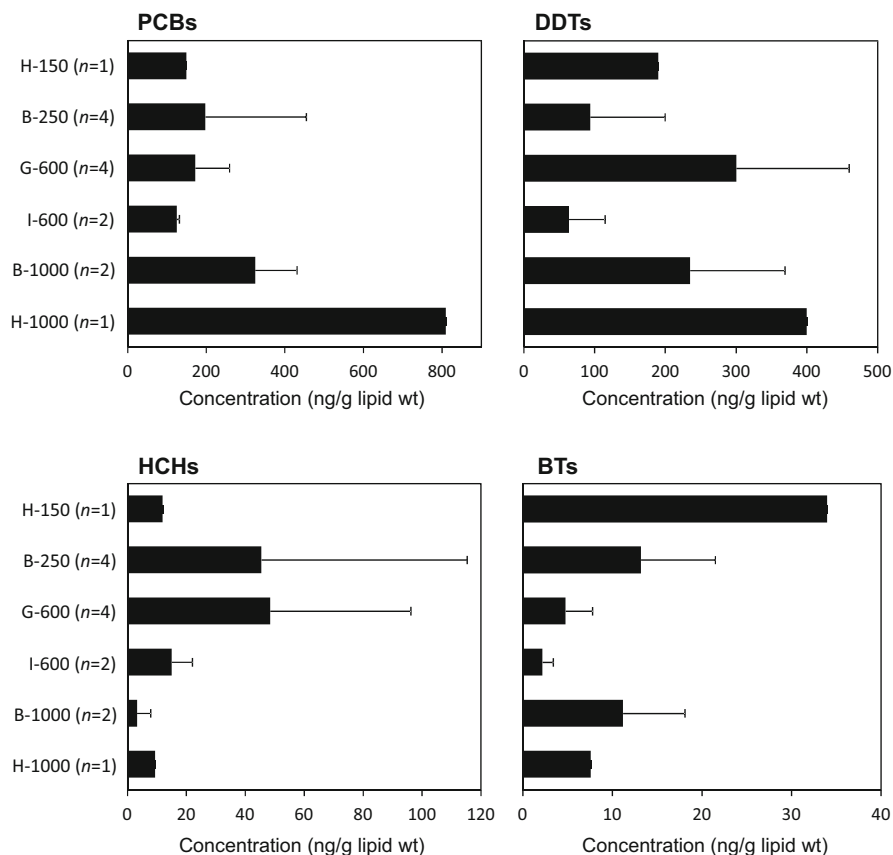
Fig. 8 Mean concentrations and residue patterns of PCBs, DDTs, HCHs, and BTs in deep-sea fishes (except for some high trophic benthic fishes) from the western North Pacific, off-Tohoku, with different sampling depth (*error bars* show standard deviations)

NPIW occur. 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.

3.4 Compositions and Temporal Trends of Organohalogen Compounds

Influence of the water-mass structure and material transport around the Okhotsk Sea and Oyashio current on the vertical profiles of OCs can be also found in the compositions of DDTs in deep-sea fishes from the WNP, off-Tohoku. Among the DDT compounds, *p,p'*-DDE and *p,p'*-DDT were the predominant constituents in
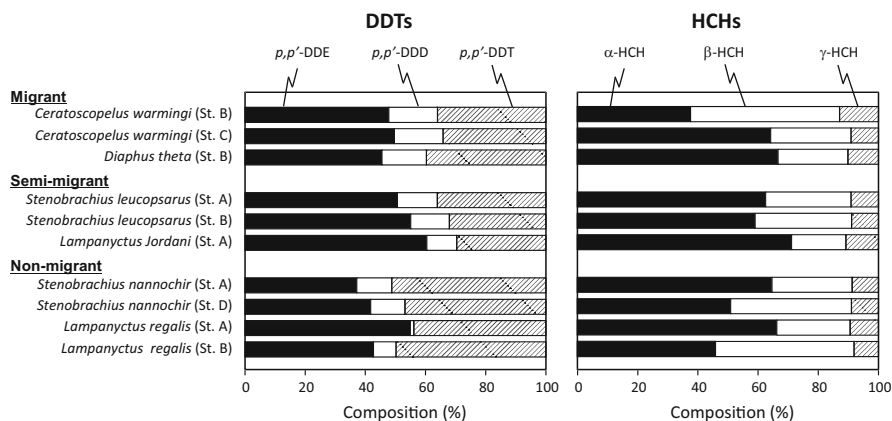
Fig. 9 Compositions of DDT compounds and HCH isomers in myctophid fishes from the western North Pacific, off-Tohoku. Each *bar* indicates the composition in a composite sample which was prepared from pooled and homogenized individual specimen belonging to the same species collected from the same sampling location [14]

almost all the deep-sea fishes (Figs. 9 and 10) [14, 44]. Similarly high proportions of *p,p'*-DDT (> 40%) were also found in Japanese common squid (*Todarodes pacificus*) from off-Tohoku, Japan, although the location is far away from any possible sources of DDT [82]. Such DDT composition (i.e., high proportions of a parent compound, *p,p'*-DDT) observed in fishes and squids from the WNP, off-Tohoku, is apparently different from those reported in previous studies from Suruga and Tosa Bays [12, 15] and in other recent researches from the Mediterranean Sea [25, 26] and North Atlantic Ocean [20, 27], where *p,p'*-DDE, a stable degradation compound, was dominant among DDTs in almost all the fishes analyzed. In this context, the intermediate water in the study area can be expected to have relatively fresh input of DDTs. Particularly in myctophids, higher proportions of *p,p'*-DDT were observed in nonmigratory fishes than in those of migratory ones ($p < 0.05$, Mann–Whitney U-test) (Fig. 9). This agrees with the concentration profile of DDTs as noted above.

Among the HCHs isomers, α-HCH was the predominant isomer in all the deep-sea fishes in the NWP, off-Tohoku (Figs. 9 and 10). It has been reported that the ratio of α-HCH to total HCH concentrations in seawater and fish has a tendency to increase with the increasing latitude [30, 63]. α-HCH is preferentially transported to northern colder regions due to higher vapor pressure among the HCH isomers.

Regarding to the temporal change in the composition of OCs, the percentages of *p,p'*-DDT in myctophids collected in 1994 (~40%) was apparently lower than those in myctophids collected from Yaizu, Suruga Bay, in 1976 (58%) [83]. This suggests the reduction of fresh input of DDTs in the WNP during the 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 [84]. In contrast to the composition of DDT compounds, no significant
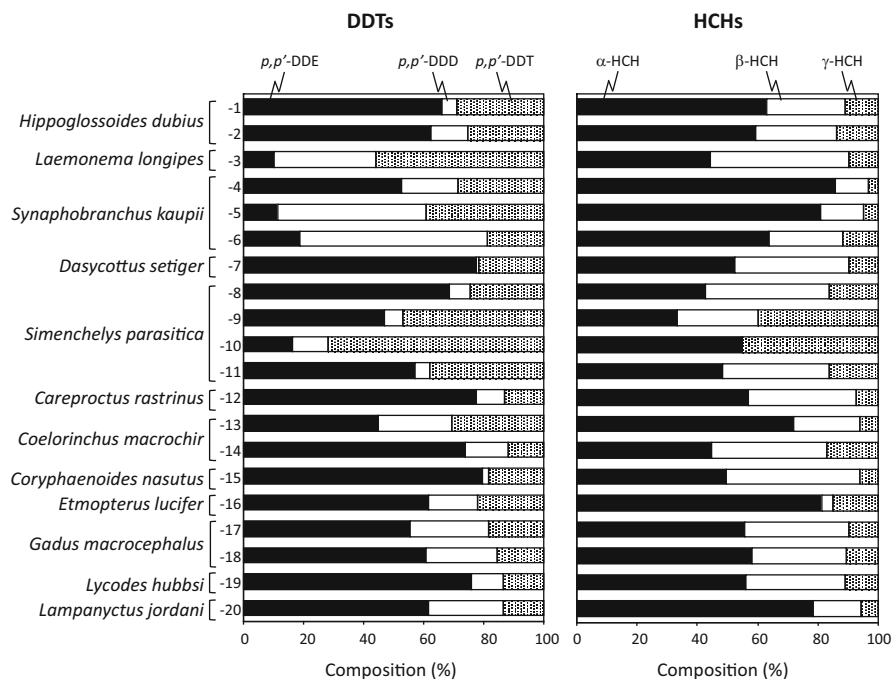
Fig. 10 Compositions of DDT compounds and HCH isomers in various deep-sea fishes from the western North Pacific, off-Tohoku

difference in the proportion of α-HCH (~60%) was observed between the myctophids collected in 1976 and 1994. Such a small variation in the temporal trend of HCH isomer composition has also been reported in the northern fur seals [84]. It may be attributable to the global transport of HCHs to high-latitude regions and/or the water-mass inflow from the Okhotsk Sea. Further monitoring along the Kuroshio–Oyashio interfrontal zone including marginal seas such as the Okhotsk Sea with geophysical and biogeochemical studies is required to delineate the distributions and fate of anthropogenic contaminants.

4 POPs and Related Compounds in the East China Sea

In this section our recent studies in the ECS [42, 43] have been reviewed for understanding the distribution and the specific accumulation characteristics of organohalogen and butyltin compounds in various deep-sea organisms (e.g., deep-sea eels, sea perches, hairtails, argentines, grenadiers, splitfins, flounders, gurnards, myctophids, lantern sharks, dogfish sharks, skates, squids, shrimps, prawns, lobsters, crabs, sea anemones; see the above references for more details).

4.1   Contamination Status

Among the organohalogen compounds analyzed in deep-sea organisms from the ECS, the concentrations of DDTs were the highest and those of the other compounds were approximately in the order of PCBs > CHLs > HCB > HBCDs > PBDEs = HCHs [42, 43] (Table 3). Deep-sea fishes from the ECS had the highest concentrations of DDTs among the data reported so far from our group for the WNP region (Table 1). Rapid industrial development and population growth in coastal areas of China have resulted in significant environmental pollution and damage to the aquatic ecosystems. From the 1950s to the 1980s, DDT was widely used in agriculture in China before it was legally banned in 1983 [85]. In the last two decades, many agricultural lands in China have been developed for commercial uses, thus, accelerating the remobilization of previously buried insecticides/pesticides. Yuan et al. [86] reported that the large-scale usage of DDT in agricultural practices and the subsequent runoff into the waterways have resulted in the high levels of DDTs in the freshwater, estuarine, and marine environment of China. In the case of PCBs, the concentrations in deep-sea fishes from ECS were comparable to that of deep-sea fishes from Tosa Bay [15] and off-Tohoku, Japan [14, 41, 44], but were significantly lower than that of deep-sea fishes from Suruga Bay [12] and other locations of the world (Table 2). On the other hand, the concentrations of HCHs and HCB in deep-sea organisms from the ECS were about one or two orders of magnitude lower than those of DDTs and PCBs. HCH isomers are less lipophilic when compared to other OCs, and, thus, they have lower biomagnification factors in aquatic ecosystems [87]. Significant fluxes (i.e., volatilization) of more volatile POPs, HCHs and HCB, to the atmosphere in the waters of low-latitude regions [2, 30, 64] may also be attributable to their less contamination in the ECS along the Kuroshio Current.

Unlike PCBs and DDT, which are largely a legacy of the past, BFRs such as PBDEs and HBCDs have been banned from usage since late 1990s–2000s or even currently used in various electronic devices, furniture and textiles. Being additive flame retardants they can leak out of the treated materials during the life cycle of the product, causing a continuous contamination of the environment even after the regulations on their production/application were implemented. Total concentrations of PBDEs and HBCDs in various deep-sea organisms from the ECS ranged from 0.31 to 57 ng/g lipid wt. and 0.15 to 210 ng/g lipid wt., respectively (Table 3). Reports on PBDEs and HBCDs in marine ecosystems and food webs, particularly for the deep-sea environment, are relatively scarce. The concentrations of PBDEs in deep-sea fishes from the ECS were higher than those from the WNP, off-Tohoku [44]. The detection of PBDEs and HBCDs in deep-sea organisms from the ECS indicates that these compounds are also transportable to the deep oceans, similar to that of other POPs. The sources of these BFRs in the ECS could be from the various manufacturing operations as well as e-waste recycling activities along the Chinese coast [88]. The recycling of e-wastes could mobilize these BFRs from the electronic components into the environment.

Table 3 Mean and range concentrations of organohalogen compounds (ng/g lipid wt) in deep-sea organisms from the East China Sea

Species	n	Fat (%)	PCBs	DDTs	CHLs	HCHs	HCB	PBDEs	HBCDs
Fishes	30	2.5 (0.48–9)	290 (36–1,400)	560 (<3.0–2,200)	51 (8.4–91)	11 (2.2–41)	23.0 (<1.5–290)	10.0 (0.31–40)	4.0 (0.25–20)
Cephalopods	3	2.1 (0.69–4.5)	120 (29–190)	220 (76–410)	39 (3.9–640)	5.7 (3.7–7.3)	8.5 (3.2–14)	11 (0.65–26)	71 (0.18–210)
Crustaceans	9	3.1 (0.47–12)	230 (25–850)	610 (21–2,500)	40 (7.5–140)	14 (4.5–28)	7.5 (0.55–17)	16 (1.1–57)	16 (0.15–73)
Mollusk	1	0.23	220	300	23	33	17	4.7	3.1
Echinoderm	1	0.041	590	72	1.9	3.4	<8.8	28	11

Figures in parentheses indicate the range of concentrations

n: number of species

For calculation of mean, data less than detection limits was assumed to be 0

The contamination status of organohalogen compounds in the ECS is probably affected by the various hydrological processes, notably the Yangtze River discharge and the Kuroshio Current. The Yangtze River flowing through big cities and important agricultural areas with an annual discharge of more than 9.0×10^{11} m^3 [89] is a significant source of anthropogenic pollutants from the terrestrial environment to the ECS. On the other hand, the Kuroshio, flowing to the northeast along the continental slope, is a unique source of oceanic materials of open-ocean origin to the shallow continental shelf [90].

Concentrations of butyltin compounds (BTs) and other organotin compounds including triphenyltin (TPT) were reported by Tanabe et al. [42]. BTs and TPT were detected in almost all deep-sea organisms analyzed (levels up to 62 ng/g wet wt in blackbelly lantern shark, *Etmopterus lucifer*, for $\sum$BT, and 40 ng/g wet wt in garden eel, *Conger myriaster*, for TPT). Although the magnitude of contamination by BTs in ECS seems to be relatively low comparing with that of the western North Pacific, off-Tohoku, the results indicate widespread contamination by organotin compounds including TPT in various deep-sea ecosystems around the East Asian region.

4.2 Species-Specific Accumulation and Composition of Organohalogen Compounds

The concentrations of most of the organohalogen compounds in deep-sea organisms from the ECS varied considerably between the various species analyzed. Factors like food web characteristics, habitat, and feeding strategies can contribute to the exposure of deep-sea organisms to organohalogen compounds. Higher concentrations of organohalogen compounds were found in fishes like garden eel (*Conger myriaster*), Japanese split fin (*Synagrops japonicus*), lantern shark (*Etmopterus lucifer*), dogfish shark (*Squalus japonicus*), sharpnose sevengill shark (*Heptranchias perlo*), and green eyes (*Chlorophthalmus acutifrons*). All these species are deep-sea species and feed at high trophic levels of the food web. Thus, the species-specific differences in the accumulation patterns of the organohalogen compounds observed are due to biomagnification and/or feeding habits of the organisms. In general, HCHs and HCB, which have lower molecular weights and are less lipophilic than the other organohalogen compounds, exhibited smaller variation in concentrations among species and trophic levels owing to their faster rates of attaining equilibrium with water concentrations [83, 91]. However, unicorn grenadier (*Caelorinchus productus*) showed very high levels of HCB (up to 290 ng/g lipid wt.). The other grenadier species from the ECS also showed elevated levels of HCB. High levels of HCB were also found in the same species from Tosa Bay, Japan [15], and off-Tohoku, Japan [41]. So far, it is not clear why grenadier species accumulate such high levels of HCB.

Among the DDT compounds, *p,p'*-DDE or *p,p'*-DDT were the predominant constituents in almost all the deep-sea organisms. The percentages of *p,p'*-DDT in shallow-water and deep-sea fishes, 37% and 40%, respectively, were higher than in
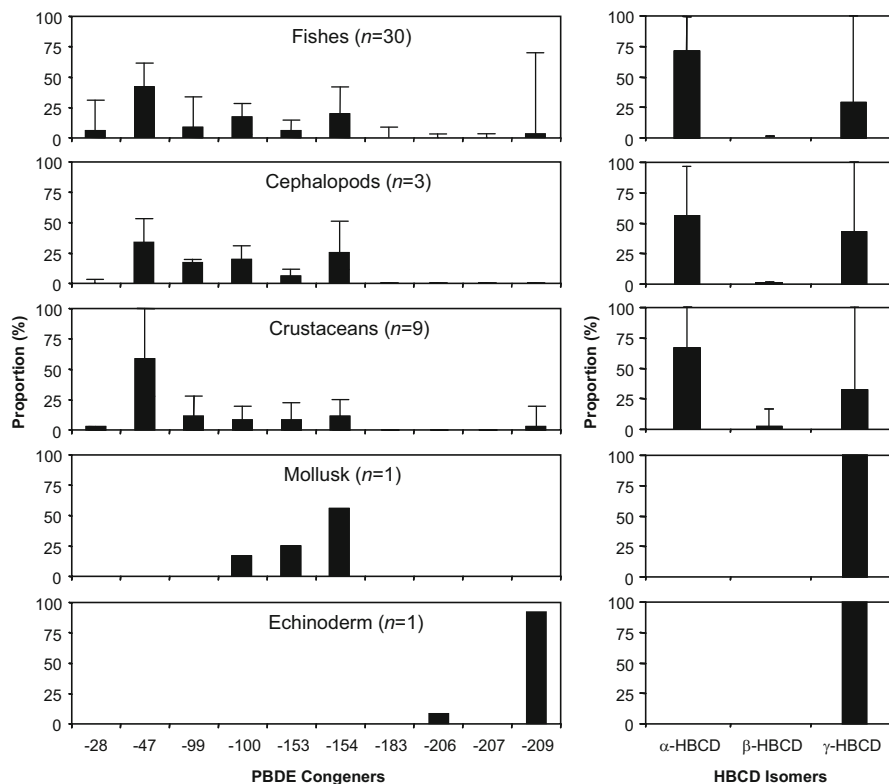
Fig. 11 Mean proportions of PBDEs and HBCDs in deep-sea organisms from the East China Sea (*error bars* show standard deviations)

the deep-sea fishes from Suruga Bay and Tosa Bay [12, 15] and other locations of the world such as the Mediterranean Sea [25, 26] and North Atlantic Ocean [20, 27], indicating that there is fresh input of DDT from China or other Southeast Asian countries to the ECS. Higher percentages of *p,p'*-DDT were also found in skipjack tuna (*Katsuwonus pelamis*) from the ECS [63]. In case of HCH composition, β-HCH was the dominant isomer in all the deep-sea organisms, and this may be because of its persistent nature. α-HCH which is the major isomer in technical HCH mixture was detected in most of the samples, but the percentage composition of α-HCH was far less when compared to deep-sea fishes from the WNP, off-Tohoku [14, 41, 44]. In spite of the usage of technical HCH mixture in China and nearby countries, the low levels of α-HCH may be due to the rapid volatilization of α-HCH in the lower latitudes [30].

Of the fourteen PBDE congeners analyzed, a total of ten congeners from tri- to deca-BDE were identified in the deep-sea organisms from the ECS. The congener profiles of PBDEs varied between the species and, in most cases, the congeners from tetra- to hexa-BDEs were predominant (Fig. 11). The higher brominated

congeners from octa- to deca-BDE were detected in very few samples, suggesting the relatively low bioaccumulation potential as well as the low bioavailability of these congeners. Congener-specific biotransformation of PBDEs undoubtedly plays an important role in the bioaccumulation behavior of PBDEs. BDE 209 was occasionally present at concentrations above its detection limit. It is noteworthy that BDE 209 was the dominant congener in case of an echinoderm (*Holothuroidea* sp.) and in flatfish (*Chascanopsetta lugubris*), and this can be explained by their sediment-dwelling nature. BDE 209 is mainly associated with sediments and is a major congener in suspended particles and sediments and related environments. The low detection of BDE 209 in other deep-sea organisms could be due to either its large molecular size which lowers its uptake rates or its relatively rapid biotransformation [92]. However, the presence of BDE 209 in deep-sea organisms from the ECS indicates that this congener probably is capable of long-range transport and dispersal. Among the HBCD isomers, α- and γ-HBCDs were detected in all the samples, while β-isomer was below detection limits in most of the samples (Fig. 11). Similar to BDE 209, higher proportion of γ-HBCD was observed in sediment-dwelling organisms.

4.3 Trophic Magnification and Sources of Contaminants in the Food Web

As noted above in the section for the WNP, the quantification of both stable carbon and nitrogen isotope ratios can provide valuable information on the feeding ecology of biota and its potential influence on the trophic enrichment of contaminants. [43] examined $\delta^{15}N$ and $\delta^{13}C$ in deep-sea organisms from the ECS to assess trophic magnification of organohalogen contaminants in the food web of the ECS. The $\delta^{15}N$ values in the deep-sea organisms from the ECS ranged from 7.18‰ to 14.17‰. No clear relationships were observed between $\delta^{15}N$ (‰) and concentrations (both in wet and lipid wt) for all the contaminants. In the aquatic environment, the uptake of contaminants by biota usually includes direct partitioning from the surrounding environment and ingestion of contaminated dietary sources, whereas the loss processes include direct partitioning, biotransformation, growth dilution, maternal transfer to offspring, and egestion. Besides, intensive biological N_2 fixation by cyanobacterium, *Trichodesmium*, which plays an important role of primary production in the ECS, can affect variation of $\delta^{15}N$ in individual organisms and entire food web [93]. The large difference in nutrient and organic matter sources between the sampled locations and specimens can be plausible reasons for the lack of relationship between $\delta^{15}N$ (‰) and concentrations of organohalogen compounds in the ECS food web, as will be explained later.

Generally, the terrigenous organic matter is depleted in ^{13}C, while the marine organic matter is usually enriched in ^{13}C [94, 95]. Since the $\delta^{13}C$ of marine planktonic sources is different from those of most terrestrial organic material sources, the carbon isotopic composition can be used as a tracer to distinguish
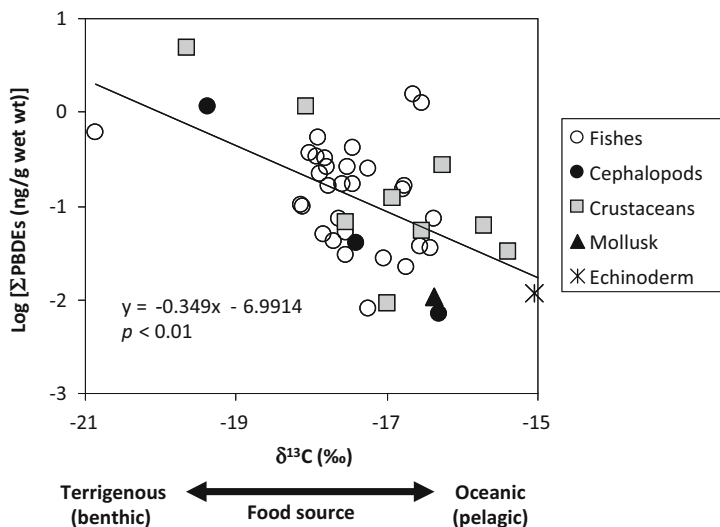
Fig. 12 Relationship between $\delta^{13}C$ (‰) and total PBDE concentrations in deep-sea organisms from the East China Sea

marine from terrigenous organics. In the present study, the $\delta^{13}C$ values in the deep-sea organisms from the ECS ranged from -15.54‰ to -20.87‰. Zhou et al. [96] reported the mean $\delta^{13}C$ values of suspended particulate matter (SPM) in the Changjiang Estuary to be -22.6‰. Therefore, the deep-sea organisms from the ECS with low $\delta^{13}C$ (-20.87‰) could indicate the input of terrigenous materials to the ECS. In the present study, strong negative relationships were observed for log-normalized lipid weight concentrations and $\delta^{13}C$ (‰) for all the analyzed organohalogen compounds (Fig. 12, data shown only for PBDEs). Thus, the negative relationships observed between the organohalogen concentrations and $\delta^{13}C$ (‰) indicate that organisms showing the higher concentrations than the other species accumulate these compounds from terrigenous (coastal benthic) sources rather than oceanic (pelagic) organic matter. Such specific pattern also reflects that steep slope in the concentrations of nutrients, terrigenous materials, and, possibly, contaminants from the coastal front zone is affected by Yangtze River discharge to oceanic waters along the Kuroshio Current.

4.4 Distribution and Transport of POPs into Deep Waters

Apart from the species-specific accumulation of organohalogen compounds, the variation in the concentrations of the organohalogen compounds among the organisms could be related to the distribution of these compounds in the marine ecosystem. Even among the fishes showing similar feeding habits, considerable variations
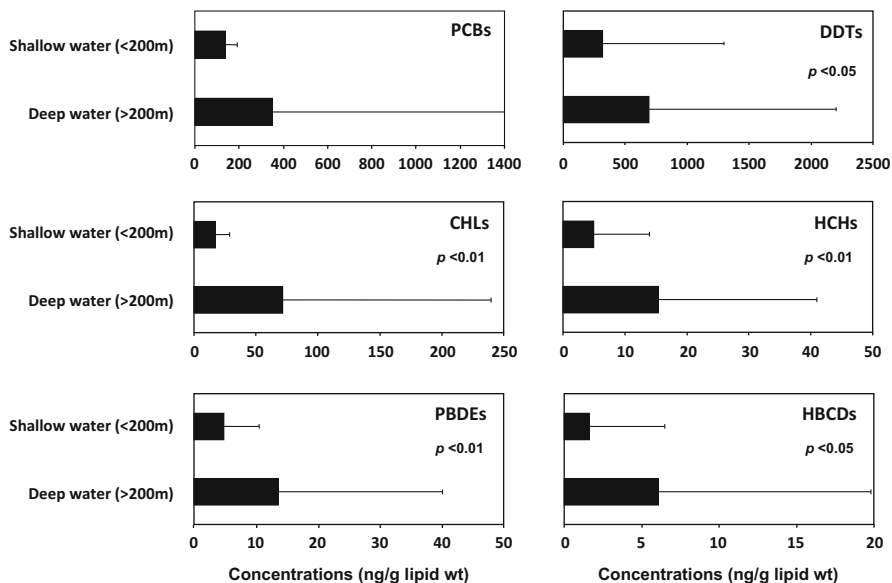
Fig. 13 Mean and range concentrations of organohalogen compounds in fishes collected from shallow (<200 m) and deep waters (>200 m) in the East China Sea (*error bars* show the maximum concentrations)

in the concentrations of organohalogen compounds were observed, which cannot be explained only by their feeding habits and food web magnification. To understand spatial and vertical distribution of the contaminants in the ECS, the fishes were grouped into two categories, shallow water (<200 m) and deep water (>200 m), based on their sampling depths. Interestingly, it was found that the concentrations of almost all the contaminants (except for PCBs) were significantly higher in fishes from greater depths than that from shallower depths ($p < 0.05$ for DDTs and HBCDs and $p < 0.05$ for CHLs, PBDEs, and HBCDs, Mann–Whitney U-test) (Fig. 13).

As it was mentioned before, the ECS shelf receives large amounts of freshwater, suspended matter, and nutrients with the Yangtze River discharge [62]. Furthermore, Gong et al. [97] reported that the ECS has a moderately high primary production (108–997 mg/m^2/d with a mean value of 425 mg/m^2/d). Thus, the high primary productivity and input of terrestrial materials with the Yangtze River discharge leads to an increase in the particulate matter. Kao et al. [61] tracked the export flux of organic matter from the shelf to the deep-sea by comparing carbon and nitrogen isotopic characteristics of sedimentary organic matter all over the ECS and the adjacent Okinawa Trough. The study found similar $\delta^{13}C$ and $\delta^{15}N$ values between sediments from the inner shelf near the China coast and from the Okinawa Trough, suggesting that the ECS shelf is a source of particulate matter to the deeper waters of the ECS.

In the marine environment, organohalogen compounds are either dissolved in the water phase or associated with suspended particulate matter or dissolved organic carbon depending on their hydrophobicity and organic carbon–water partition coefficients (K_{OC}). The particulate matter, on which POPs sorb by the process of flocculation and aggregation to be eventually deposited into the deep waters, could be of natural origin, like detritus, fecal pellets, and living planktonic organisms, or from an anthropogenic source. Studies have shown that eutrophication results in large vertical fluxes of particle-associated pollutants and organic carbon to the deep waters [38, 52]. Thus, all the examined organohalogen compounds being hydrophobic are expected to have high affinity to particulate matter and get transported to deep waters. Volatilization of semi-volatile compounds like HCHs and HCB to the atmosphere from the surface waters of ECS due to the high temperatures in the subtropical region could also contribute to the vertical gradient of their concentrations. On the other hand, biological and ecological factors which can influence the results obtained from various organisms should be considered for further discussions on behavior of POPs. It is necessary to develop a useful model including key biological and ecological parameters for further "normalization" of the contaminant concentrations in organisms (instead of simple lipid normalization) as well as to use better "bioindicators" comprising of same/similar species for future research.

5   Conclusions and Perspectives

It is noteworthy that higher concentrations of POPs and related compounds in deeper waters were indicated by using bioindicators such as fishes in the WNP, off-Tohoku, and ECS, which have been documented to be high productive frontal zones around East Asia. Expansion of contamination by emerging POPs including PBDEs into the deep-sea ecosystems was also suggested by our research. Our recent ocean survey on POPs in sediments from the ECS reported significantly higher concentrations of DDTs and PCBs in deep-sea sediment from the Okinawa Trough than those in the shallower continental shelf [98]. These results suggest the vertical transport of POPs and related compounds in the high productive waters along the oceanic fronts and the potential role of deep-sea bed as a final sink and reservoir for these persistent contaminants. The accumulation and distribution of the POPs and related compounds observed in the marine ecosystems reflects their physicochemical properties such as vapor pressure and hydrophobicity as well as biological and ecological factors of organisms. In addition, riverine discharges, advection by oceanic currents, and formation of oceanic fronts are some examples of the physical transport processes that influence the redistribution of POPs. Further interdisciplinary approach including studies on biogeochemical and geophysical processes in the ocean as well as field observations are required to delineate the global and regional fate of POPs.

Mathematical models are one of indispensable tools to describe the chemodynamics of POPs, including the mass budget, half-life, and long-range transport potential. Models

have been used to study the role of atmospheric transport in the multi-compartmental global fate and distribution of persistent substances. However, high-resolution global models that predict nonsteady dynamics of POPs in oceans need to be developed to understand the potential impact of POPs on ecosystems, human health, and their ultimate fate. In this regard, our group has developed a model called the Finely Advanced Transboundary Environmental model (FATE) [99, 100]. The FATE is a global high-resolution multi-compartment model that computes nonsteady POPs biogeochemical cycles in and across the five environmental compartments (atmosphere, ocean, vegetation, soil, and cryosphere). This model enables to predict the three-dimensional distributions of POPs as a result of advective transports, and to quantify the amount of PCB exports to the deep ocean by several physical and biogeochemical processes including phytoplankton detritus settling. Furthermore, we have developed a three-dimensional high-resolution transport model for regional scale POPs fate in the ECS [101], which will be linked and evaluated by our POP observations in the ECS for three consecutive years from 2008 to 2010 [98]. Using such state-of-the-art models, we were able to predict global and regional sinks and reservoirs of POPs which could provide new insights into our current understanding of the fate and transport of POPs.

Recently, we have also started looking at the chemical aftermath of the Tohoku earthquake (the Great Eastern Japan Earthquake) and tsunami that took place on March 11th, 2011 [102]. Three nuclear reactors at Fukushima Daiichi Nuclear Power Station in the tsunami area suffered hydrogen explosions, and leaked radioactive substances including 29.3 PBq of ^{137}Cs were estimated to be released into the ocean [103]. These and other radioactive materials are now polluting the global environment, and it has been cautioned by many that these may accumulate in the biotic compartments and may ultimately reach human. This phenomenon needs the maximum attention of scientists working on the aftereffects of the Great Eastern Japan Earthquake. In addition, the tsunami had wiped out a strip of coast supporting a wide range of land uses and industries. Petro- and agrochemical plants, iron foundries, steel works, and automotive, electronics, food processing, paper, plastics, and pharmaceutical plants were among those that suffered damage. Numerous reports available indicate that various chemicals with potential environmental and health hazards may have been present at many facilities in heavily impacted locations and that old electrical equipment containing PCBs was washed away [104]. The complexity of the waste generated during the Great Eastern Japan Earthquake and tsunami may pose a serious threat of environmental pollution, which should also be monitored along with the effects of nuclear disaster. Our previous study [105] showed that the levels of ^{137}Cs was the lowest in the species of marine mammals obtained from off-Tohoku, Japan, when compared with the specimens caught from other parts of the world such as Lake Baikal, Black Sea, North Sea, and Northern Canadian waters. As reviewed in this chapter, the data on POPs and related compounds in various organisms in the WNP, off-Tohoku, Japan, was also based on the observations in the region where the Great Eastern Japan Earthquake occurred. Thus, we would like to reiterate here that work on POPs and other emerging contaminants (e.g., brominated flame retardants, perfluoro-chemicals, pharmaceutical and personal care products, toxic elements) as well as

radionuclides on the specimens from this region now can give an insight into the most discussed environmental problem in the area. The papers reviewed here can provide the baseline data for comparison for studying the possibility of build-up of the chemical contaminants in the marine ecosystems along northeastern Japan. We hope that our outcomes and future trials on the marine pollution can contribute to avoid any possible catastrophe by such chemicals around our country and the world.

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