Selection of Bioindicators of Polybrominated Diphenyl Ethers, Polychlorinated Biphenyls, and Organochlorine Pesticides in Mollusks in the Chinese Bohai Sea

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Mollusks, including bivalve and gastropod samples, were collected in the summer of 2006 and 2007 to evaluate the spatial patterns of organochlorine pesticides (OCPs), polychlorinated biphenyl (PCBs), and polybrominated biphenyl ethers (PBDEs) in the coastal water of the Chinese Bohai Sea. Seven species of mollusks, including *Rapana venosa* (Rap), *Neverita didyma* (Nev), *Scapharca subcrenata* (Sca), *Mytilus edulis* (Blue mussel, Myt), *Amusium* (Amu), *Meretrix meretrix* (Mer), and *Crassostrea talienwhanensis* (Oyster, Ost) were investigated to select appropriate bioindicators. Compared with other species, Ost and Myt possess higher lipid contents and have higher concentrations of OCPs and PBDEs. Multivariate linear regression analysis revealed that the most influential factor on the contaminant concentrations in mollusks is the trophic level (TL), followed by the lipid contents. Concentrations of OCPs, PCBs, and PBDEs correlated negatively with the TL of the selected mollusks. Results of principal component and correlation analyses suggest that Ost and Myt can be used as potential bioindicators of pollution by OCPs, PCBs, and PBDEs of the Chinese Bohai Sea.

Introduction

Persistent organic pollutants (POPs) are of great environmental and health concern. Several organochlorine pesticides (OCPs) and polychlorinated biphenyls (PCBs) have been targeted for restriction or ultimate elimination by the Stockholm Convention on POPs which went into effect in 2004. Other groups of organic contaminants, including polybrominated diphenyl ethers (PBDEs), are considered the next generation of POPs to be included into the treaty due to their similarities to the banned chemicals in regard to health effects as well as their ubiquitous presence in the environment (1).

OCPs were widely used in China from the 1950s to the 1980s. Although the usage of OCPs was banned in 1983,

evidence shows that some OCPs, such as lindane (γ -HCH), are still being used in the country (2). An estimated 10 000 tons of PCBs were manufactured in China between 1965 and 1974 and were used primarily as an additive in paint and as a dielectric fluid in transformers (3). PBDEs have been found to severely contaminate several coastal areas of China, which is mainly attributed to the electronic waste import and dismantling and the manufacturing of decabromodiphenyl ether (1, 3).

To elucidate the distribution of POPs on global and regional scales, aquatic or marine organisms have been used as bioindicators (4–6). Different from other species (e.g., skipjack tuna and snapping turtles) used previously (5, 6), mollusks have unique characteristics. As water-respiring filter feeder species with a sessile lifestyle, their elimination of persistent pollutants with $K_{ow} > 10^5$ is sufficiently slow, which causes bioaccumulation of these chemicals (7). Mollusks including bivalves and gastropods have long been used as potential bioindicators and biomonitors, (e.g. in the Mussel Watch program (8)) for their high accumulation capacity and abundance in marine ecosystems. In the Asia–Pacific mussel watch program, Tanabe and co-workers (9–11) assessed contamination by various POPs using mussels in the coastal waters of Asian countries, including China, India, Japan, Korea, and more.

The Bohai Sea of China is surrounded by four provinces, with a number of large cities near its shoreline. Historically over nearly four decades, large amounts of OCPs were manufactured and applied for agricultural use in the region. With about one-sixth of the total population and a quarter of the total GDP of China, the Bohai Bay Rim economic zone has now become a hot spot for investment. However, the rapid socioeconomic growth has raised concerns about the potential deterioration of the Bohai ecosystem.

This work is a comprehensive investigation of OCPs, PCBs, and PBDEs in mollusks collected from coastal water of the Chinese Bohai Sea in 2006 and 2007. The objectives are to select the most suitable mollusk species as bioindicators and to examine the temporal trend and spatial distributions of these pollutants in the area.

Experimental Section

Sampling. Mollusks were collected from nine coastal cities along the Bohai Sea, including Dalian (DL), Yingkou (YingK), Huludao (HLD), Qinhuangdao (QHD), Tianjin (TJ), Yangkou (YangK), Penglai (PL), Yantai (YT), and Weihai (WH). A map showing sampling locations is given in the Supporting Information (Figure S1).

The sampling was carried out in late July to early August of 2006 and 2007. A total of 114 samples (59 in 2006 and 55 in 2007) were collected. Seven species of mollusks, including *Rapana venosa* (Rap), *Neverita didyma* (Nev), *Scapharca subcrenata* (Sca), *Mytilus edulis* (Myt), *Amusium* (Amu), *Meretrix meretrix* (Mer), and *Crassostrea talienwhanensis* (Oyster, Ost) were identified (Table S1) (12). Detailed sample collection and transportation procedures have been described previously (13, 14).

Sample Analysis. Sample pretreatment, instrumental analysis, quality assurance and quality control, and quantification of the data followed our previously established methods (13, 14), with minor modifications, and are described in detail in the Supporting Information. OCPs, including hexachlorobenzene (HCB); α -, β -, γ -, and δ -hexachlorocyclohexanes (HCH); *o,p'*-DDE; *p,p'*-DDE; *o,p'*-DDD; *p,p'*-DDD; *o,p'*-DDT; and *p,p'*-DDT, were analyzed using Agilent 6890A gas chromatography equipped with a ^{63}Ni electron

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TABLE 1. Concentrations of OCPs, Σ PCBs, and Σ PBDEs in Mollusks from the Chinese Bohai Sea (ng/g, dw)

		2006 ^a					2007				
		mean	median	min	max	n	mean	median	min	max	n
Mer	HCB+HCH	15.5	13.4	8.44	31.2	9	16.0	8.52	3.69	62.7	7
	Σ DDT	43.4	30.3	2.82	139	9	88.9	40.6	19.0	388	7
	Σ PCB	1.76	1.15	0.67	4.28	9	1.80	1.21	0.59	5.71	8
	Σ PBDE	1.99	1.44	0.22	5.88	8	0.61	0.45	0.04	1.94	8
Ost	HCB+HCH	8.82	7.31	5.36	7.95	8	3.48	3.23	bdl	9.88	8
	Σ DDT	260	284	39.6	521	8	417	461	165	578	8
	Σ PCB	3.42	2.00	0.84	11.5	9	3.45	3.55	1.74	5.46	8
	Σ PBDE	2.52	2.41	0.51	5.19	9	1.74	1.98	0.28	2.42	8
Amu	HCB+HCH	9.39	5.36	4.54	26.6	7	1.06	bdl	bdl	6.16	8
	Σ DDT	142	133	66.2	331	7	117	111	81.4	179	8
	Σ PCB	1.75	1.24	1.09	3.93	8	2.04	2.10	0.59	3.14	8
	Σ PBDE	1.46	1.53	0.88	2.02	8	1.08	1.21	0.07	1.87	8
Nev	HCB+HCH	3.29	2.96	0.56	7.46	10	0.67	bdl	bdl	2.63	7
	Σ DDT	38.6	30.5	14.5	102	10	38.4	36.2	17.0	72.2	7
	Σ PCB	7.78	1.45	0.34	29.9	10	4.29	1.91	0.93	11.9	7
	Σ PBDE	0.69	0.66	0.11	1.58	9	0.99	0.56	0.26	2.55	7
Rap	HCB+HCH	5.14	5.63	0.56	9.89	8	1.42	bdl	bdl	6.71	9
	Σ DDT	110	100	1.79	204	8	75.4	73.0	10.9	204	9
	Σ PCB	1.73	1.53	0.59	2.74	9	2.03	2.08	0.63	3.06	9
	Σ PBDE	2.01	1.33	0.14	5.71	9	0.93	0.71	0.38	2.68	9
Myt	HCB+HCH	14.6	11.2	6.67	34.1	7	8.18	6.68	bdl	25.1	7
	Σ DDT	191	203	151	227	7	156	89.9	24.4	416	7
	Σ PCB	1.84	2.04	0.38	2.77	8	3.75	2.02	1.24	9.60	7
	Σ PBDE	32.6	51.1	0.88	60.9	7	1.70	1.24	0.50	4.39	7
Sca	HCB+HCH	20.3	9.04	5.30	99.5	9	3.77	2.90	bdl	13.1	10
	Σ DDT	103	68.3	16.9	293	9	95.0	61.2	7.95	324	10
	Σ PCB	1.55	1.31	0.99	3.22	9	1.50	1.56	0.46	2.68	9
	Σ PBDE	7.52	0.75	0.33	59.0	9	0.99	0.78	0.23	3.15	8

^a Most water content of mussel samples ranged from 80 to 90%, except for some individual samples that were less than between 70 and 80%.

capture detector. PBDEs and PCBs were analyzed by the isotope dilution method using Agilent 6890 gas chromatography coupled with Waters Micromass high-resolution mass spectrometer. Fourteen PBDE congeners (BDEs 17, 28, 47, 66, 71, 85, 99, 100, 138, 153, 154, 183, 190, and 209) and 25 PCB congeners, including 12 coplanar congeners (CBs 77, 81, 105, 114, 118, 123, 126, 156, 157, 167, 169, and 189), six indicator congeners (CBs 28, 52, 101, 138, 153, and 180), and other congeners (CBs 3, 15, 19, 202, 205, 208, and 209), were analyzed. A batch of 57 samples collected from 2003 to 2005 was also analyzed for PBDEs to investigate the temporal trends in recent years.

Data Analysis. Statistical analyses were performed with SPSS for Windows Release 13.0 (SPSS Inc., 1989–2006). Principal component analysis (PCA) was executed to project multidimensional data onto two or three dimensions for comparison of the congener distributions using varimax normalized rotation on an untransformed matrix. The principal components (PC) were usually obtained with their eigenvalues > 1.

Results and Discussion

Table 1 summarizes the dry-weight-based concentration sums of OCPs, PCBs, and PBDEs in mollusks. The concentrations of individual OCPs, PCBs, and PBDEs are illustrated by box-and-whisker plots in Figure 1. The spatial distributions and the composition are illustrated in Figures S2 and S3 of the Supporting Information, respectively. Concentrations of PBDEs in mollusks collected in 2003 and 2005 are shown in Figure S4 of the Supporting Information.

HCHs. The concentration of Σ HCH (sum of HCHs and HCB) in the mollusks ranged from 0.56 (Nev, TJ) to 99.5 ng/g (Sca, WH) in 2006 samples and from below detection limit (BDL) to 62.7 ng/g (Mer, TJ) in 2007. On the basis of lipid content, the concentration was in the range of 9.33 (Rap, PL)

to 829 ng/g lipid (Sca, WH), with a mean of 141 ng/g lipid in 2006, and from BDL to 1045 ng/g lipid (Mer, TJ) with a mean of 54.2 ng/g lipid in 2007. The levels of Σ HCH in mollusks are comparable to those reported by Ramu et al. (15) from this area (Dalian and Qingdao), but higher than those found in other sites in Asia, except for India and a sampling site in Japan.

The concentrations of HCHs were in the rank order β -HCH > δ -HCH > γ -HCH > α -HCH (Figure S3 of the Supporting Information). Compared with other isomers, β -HCH has lower vapor pressure and a higher bioenrichment factor and is more stable in organisms. In addition, β -HCH may be formed from other isomers in the environment. Therefore, environmental concentration of β -HCH is often higher than other isomers, despite its low relative amount (6–10%) in commercial mixtures (16). The ratio of α -/ γ -HCH was 0.76 (2006) and 0.52 (2007) in mollusks, suggesting that lindane is still being used in the Bohai Sea, which is in agreement with the conclusion of Ramu et al. (15).

DDTs. As the dominant contaminant group, Σ DDT, which includes *o,p'*-DDE, *p,p'*-DDE, *o,p'*-DDD, *p,p'*-DDD, *o,p'*-DDT, and *p,p'*-DDT, has much higher concentrations than Σ HCH in Bohai Sea mollusks. The Σ DDT ranged from 1.79 (Rap, HLD) to 521 ng/g (Ost, TJ) in 2006 and from 7.95 (Sca, BDH) to 578 ng/g (Ost, WH) in 2007. The lipid-based concentrations ranged from 35.2 (Mer, HLD) to 9302 ng/g (Rap, WH) in 2006 and from 102 (Mer, DL) to 6468 ng/g (Sca, TJ) in 2007. These concentrations were lower than those measured by Ramu et al. (15) for this area as well as for other coastal sites in China and Vietnam, but higher than those in India and Japan (15). Ramu et al. (10) reported DDT concentration in blue mussels (*M. edulis*) from coastal waters of Korea in the range of 21 and 400 ng/g (lipid), which was lower than the DDT measured in this work for the same species in the Bohai Sea. No obvious temporal trend of Σ DDT concentrations in mollusks in the

FIGURE 1. Box-and-whisker plots of HCHs, DDTs, PCBs, and PBDEs in mollusks collected from the coastal water of the Bohai Sea in 2006 and 2007. For PCBs and PBDEs, "other" represents the sum of the congeners in which the concentrations are less than 5% of Σ PCB or Σ PBDE. The horizontal black line in the box represented the median value and the low and upper edge of the box mark the 25th and 75th percentiles. The whiskers extending from the box show the highest and lowest values. "*" represents extreme values, which were beyond the triplication of the difference between 25th and 75th percentiles, and singular values are represented by "O", which were beyond the 150th percentile of the difference between 25th and 75th percentiles.

Bohai Sea was found from 2002 to 2007 by comparing the data of this work with that reported previously (13).

Composition of Σ DDT is shown in Figure S3 (Supporting Information). Concentrations of p,p' -DDE, p,p' -DDD, and

p,p'-DDT were found higher than their respective *o,p* isomers. The average ratio of $\Sigma(\text{DDE} + \text{DDD})/\Sigma\text{DDT}$ in mollusks collected in 2006 and 2007 was 7.08 (2006) and 4.81 (2007), respectively. These are much higher than the ratio of 0.5 we reported for the same area in 2002–2004 (13). DDTs can be decomposed to DDE and DDD under environmental condition. The decreasing DDT proportion in ΣDDT and comparison to the concentration trends of DDTs in mollusks from 2002 to 2007 (13) implied no recent input of DDTs to this area.

PCBs. PCB concentrations in mollusks collected from the Chinese Bohai Sea were far lower than OCP concentrations (Table 1), and varied depending on the sampling location and mollusk species. The sum of all 25 PCB congeners (ΣPCB) ranged from 0.34 (Nev, HLD) to 30 ng/g (Nev, WH) in 2006 and from 0.46 (Sca, DL) to 12 ng/g (Nev, YangK) in 2007. The lipid-based ΣPCB ranged from 4.4 (Mer, TJ) to 596 ng/g (Nev, WH) in 2006 and from 6.0 (Sca, DL) to 294 (Nev, YangK) in 2007. In general, the PCB levels in mollusks in the Bohai Sea area were at the low end of the range for Asian coastal waters reported by Ramu et al. (15).

The major PCB homologues in the mollusks were tri-, tetra-, penta-, and hexa-CBs, which accounted for 20.7, 13.9, 22.7, and 29.3% of ΣPCB , respectively (Figure S3, Supporting Information). Indicator PCBs (CBs 28, 52, 101, 138, 153, and 180) represented 58.9% of ΣPCB . A strong linear relationships was found between the sum of indicator congeners and ΣPCB ($R^2 = 0.42$, $P < 0.05$).

PBDEs. PBDEs were detected in all mollusks species. Of the 14 congeners analyzed, BDEs 190 and 209 were below the detection limits in most samples and therefore not included in the data analysis. The sum of tri- to hepta-BDEs (ΣPBDE) ranged from 0.11 (Nev, HLD) to 61 ng/g (Myt, YT) in 2006 and from 0.04 (Mer, TJ) to 4.4 ng/g (Myt, PL) in 2007. Compared with the concentrations in samples collected in 2003 and 2005 (Figure S4, Supporting Information), no obvious temporal trends of ΣPBDE were found from 2003 to 2007 in Bohai Sea mollusks with the exception of Myt.

The concentrations of ΣPBDE in mollusks were lower than ΣPCB , as evidenced by the ratio of $\Sigma\text{PCB}/\Sigma\text{PBDE}$ (median = 2 and average = 4), but no clear correlation was found between the two groups ($P > 0.05$). On the whole, PCBs and PBDEs in mollusks collected from southern sampling sites were higher than those from northern sites (Figure S2 of the Supporting Information). The spatial variability was more pronounced than that of OCPs and may reflect the higher population density and industrialization in the southern coastal cities than in the northern sites. It is also likely that the high PBDE levels in the southern sites are related to the rapid economic development during the past two decades in Shandong province, which is located in the southern Bohai Sea area and is the second largest gross domestic product contributing province in China. Compared with other areas in the world, the concentrations of ΣPBDE in Myt (mussels) from the Bohai Sea in 2006 were higher than those in Korea (17) and Europe (18, 19), but lower than that found in bivalves from the San Francisco Bay (20). PBDE concentrations in Ost were lower than those from Hong Kong, Singapore, and the Philippines (15).

BDEs 47, 99, and 100 are found prevalent in biological samples worldwide. In the present study, tri- and tetra-BDE congeners accounted for 22.7 and 50.0% of ΣPBDE for all the mollusk species. Interestingly, BDE66 was the second-most-dominant congener after BDE47 in Myt, whereas in other species such as Nev, it was below the detection limit. BDE66 could be an intermediate product of metabolic debromination of penta-BDEs (e.g., BDE99) to tri-BDEs, such as BDE28. These findings are consistent with that of another study in this area (21). Elevated fractions of lower congeners suggest the occurrence of debromination of highly bromi-

nated congeners in the mollusks (22) and confirm the prevalence of biotransformation of these compounds in marine organisms.

Congener ratio BDE99/BDE100 has been found to be distinguishably different among environmental samples. In abiotic matrixes such as air and sediment, the concentration of BDE99 is often much higher than BDE100, with their ratio close to 84:16, as in the technical penta mixture. In fish and marine mammals, the opposite is often found, with a 30:70 ratio (23, 24). In the present study, the median of this ratio is about 70:30 in all mollusks except Sca. This is close to the 80:20 in blue mussel and shrimp reported by Voorspoels et al. (24). It is interesting to see that in Sca samples, the ratio ranged from 26:74 to 37:63 (2006) and from 22:78 to 39:61 (2007), with the exclusion of three samples collected in 2006 from YT (58:42), TJ (88:12), and WH (58:42). The large difference may reflect the metabolic capacity of different marine species as well as the possibly higher biodegradability of BDE99 compared with BDE100. In selecting bioindicators, species with higher metabolism of the pollutants of interest are less desirable.

Comparison among mollusk species. Mollusks possess most of the essential characteristics of ideal bioindicators, as discussed by Tanabe and Subramanian (25). To select the most suitable, comparison among mollusk species was made on the basis of their relative accumulation potential and the correlation with the concentration data in sediment. In addition, PCA was used to provide additional information in the selection of mollusk bioindicators.

No significant difference in the concentrations of ΣHCHs was observed among selected species (Figure 1), although the medians in Mer and Myt were slightly higher than those in other species. However, significantly higher levels of ΣDDT were found in Ost than in other species. The first three PCs of PCA together represent 72.4% of the total variance in OCPs concentrations in mollusks. The loading and score of the first three PCs are shown in Figure S5a of the Supporting Information and Figure 2a, respectively. Comparing the loading and score plots of PC1 vs PC2 and PC1 vs PC3, it can be seen that Ost from different sampling sites are congregated into one group, which indicates high concentrations of DDTs. However, for HCHs and HCB, no specific species appears to be more accumulating than others, although Mer shows a slightly higher ability to accumulate HCHs and HCB.

Both Nev and Ost have higher average ΣPCBs , but only Ost has a significantly higher median than the other species (Figure 1). The first three PCs of PCA accounted for 59.1% of the variance in ΣPCB concentrations in mollusks. The plot of PC1 vs PC2 (Figure 2b and Figure S5b of the Supporting Information) revealed that Ost contained higher concentrations of hepta- and hexa-CBs, such as CBs 118, 138, and 157. Higher levels of CBs 28, 77, 126, and 169 were found in the Nev samples. The grouping of PCBs shown in the plot of PC1 vs PC3 is somewhat different from the plot of PC1 vs PC2. In the loading plot, PCB congeners were divided into three groups by their degree of chlorination. Lowly chlorinated congeners are mainly located under the *y*-axis, which are accumulated in Sca, Ost, and Myt. Highly chlorinated congeners are above the *y*-axis and mainly accumulated in Nev, Rap, and Mer. Penta- and hexa-CBs are on the right side of the score plot and mainly accumulated in Ost and Amu. Nev collected at site Weihai and Ost at sites Yingkou and Dalian show higher PCB levels than other species.

The concentrations of PBDEs in Myt (2006) were found to be much higher than other species (Figure 1). PCA results (Figure 2c and Figure S5c of the Supporting Information) show that BDEs 138 and 183 have higher loading values of PC2 in the component plot. The first three PCs account for 81.9% of the total variance. In the PC1 vs PC2 plot, strict clustering composed of Myt samples and one sample of Sca

FIGURE 2. The score plots of PCA by the database of organohalogen compounds in mollusks collected from the Chinese Bohai Sea in 2006 and 2007: (a) OCPs, (b) PCBs, (c) PBDEs.

(TJ) is shown on the right-hand side, indicating higher concentrations of BDEs 17, 28, 47, 66, 99, and 100 in these samples. High concentrations of BDEs 138 and 183 can be found in species such as Ost, Mer. In the plot of PC1 vs PC3, Myt and Ost showed higher scores than other species, suggesting their higher accumulation potential for PBDEs.

PCA was also conducted for the sum of HCHs, DDTs, PCBs, and PBDEs in mollusks collected in 2006 and 2007. The first three PCs accounted for 57.1% of the total variance. Comparing the loading and score plots (Figure 3), Ost (black dot in the left figure) was found to be more sensitive and suitable for indicating the contaminations of HCHs, DDTs, PCBs, and PBDEs in this area.

A previous study in 2006 investigated the status of PBDEs in sediments from the coastal sites including Penglai, Yangkou, Tianjin, Qinhuangdao, Huludao, Yingkou, and Dalian in the Bohai Sea (26). Using these results, we analyzed the relationships between PBDE congener concentrations in mollusks and in sediments from the corresponding sampling sites. The results shown in Figure 4 indicate that PBDE concentrations in Myt and Ost are more correlated with those in sediments than other species. The results imply

that certain mollusks do not exhibit observable metabolism of PBDEs, as high linearity was found for the correlation between PBDEs in mollusks and sediment. Therefore, we consider Myt and Ost to be considered to be more appropriate than other species in indicating pollution at the sampling sites.

Relevance to Trophic Level (TL). Organohalogen compounds are more effectively accumulated in marine organisms with a higher TL within a food web. Nitrogen isotope ratios can indicate the trophic position of a marine animal and have been widely used to study the biomagnification of organic contaminants via the food chain (14). In general, mollusks are considered as being at the second lowest TL in their marine food web. A detailed investigation of food web in the Chinese Bohai Sea found TL of bivalves and gastropods in the range of 1.91–3.32 (27). The main sources of contaminants in mollusks were from the intake of food, including phytoplanktons and zooplanktons. Considering their living habit, the levels of contaminants in mollusks may to a great extent indicate the pollution of their resident sites, if metabolism in the mollusks is not an issue of concern. In this work, nitrogen isotope ratios ($\delta^{15}\text{N}$) of the selected

FIGURE 3. Loading and score plot from PCA using the database of total OCP, PCB, and PBDE concentrations in mollusks collected from the coastal water in the Chinese Bohai Sea in 2006 and 2007.

FIGURE 4. The coefficient square distributions of PBDE congener concentrations in mollusks versus PBDE congener concentrations in the sediment of the corresponding sampling sites.

mollusks were measured, and their TL were estimated using the same method in our previous work (14). Rap had the highest TL (average value 2.84), and Mer had the widest range (Figure S6, Supporting Information); and the results are in agreement with the finding of Wan et al. (26).

Inverse relationships between the concentrations and TL were found for all three groups of organohalogen (Figure S7, Supporting Information) but the linear correlations between the TL and the concentrations of PCBs, HCHs, and PBDEs were not significant ($P > 0.05$). The slopes of the regression were steeper for less hydrophobic HCHs and DDTs than for the highly hydrophobic PCBs and PBDEs. Especially, a significant negative linear relationship exists between Σ DDT and TL ($R = -0.57$, $P < 0.05$). A recent work showed that less hydrophobic compounds ($K_{ow} < 10^5$) such as HCHs and tetra-CBs do not biomagnify in the piscivorous food web with negative correlation between HCHs and TL (7).

To analyze the factors that influence the pollutant concentrations in mollusks, multilinear regression analysis (stepwise regression) was executed for the targeted POPs against the TL, lipid contents, body size, and body weight of the mollusks. The strongest regression was obtained with TL, which showed a negative correlation, followed by lipid

contents. Neither shell size nor body weight of mollusks appeared to affect the concentrations of the contaminants.

The relationships between the biomagnifications of POPs and TL in aquatic food chain remain far from being understood. In large-scale food webs, such as a phytoplankton–zooplankton–bivalves–fish–seabird food chain, biomagnifications are always observed. However, for certain real food webs containing species with a narrow, low-TL range and containing species with relatively low metabolic capacity for the contaminants, the orderliness could be disturbed by other factors of the organisms or the partitioning behavior of the contaminants. As poikilotherms, mollusks generally exhibit lower magnification through food web than homeotherms (28). In addition, the concentration of ^{15}N in mollusks varies depending on the food supplies in the habitat and the season, both of which affect their physiology. These may weaken the simple correlation and complicate the investigation on the relationships between TL and contaminant accumulation in mollusks.

Various mollusks have different abilities to accumulate POPs. The selection of bioindicators is necessary, since there is a vast variety of mollusks in the marine ecosystem. Our present work proved that Ost and Myt are the most sensitive bioindicators among the selected species in indicating the pollution by POPs in the Chinese Bohai Sea. Multilinear regression analysis revealed that, among the measured physiological parameters, TL was most related to the concentrations of the POPs targeted in this study. Our investigation has also proved that lipid contents were higher in Ost and Myt than that in other species. We therefore suggest the use of these two species as bioindicators of organohalogen compounds, which tend to be partitioning toward equilibrium among the ambient water/sediment and the lipid pool of the mollusks during their uptake and depuration processes.

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Supporting Information Available

Additional information as noted in text. This information is available free of charge via the Internet at <http://pubs.acs.org>.

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