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Biomagnification of persistent chlorinated and brominated contaminants in food web components of the Yellow Sea

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ABSTRACT

Concentrations of polychlorinated biphenyls (PCBs), organochlorine pesticides (OCPs), and polybrominated diphenyl ethers (PBDEs) were measured in 32 species inhabiting the Yellow Sea to assess their bioaccumulation potentials. The concentrations in these samples were lower than those reported for other countries or locations. Relatively high levels of BDE 209 in biota suggest an ongoing source of deca-BDE technical mixing within the Yellow Sea. The accumulation profiles of PCBs were uniform between species, but the concentrations of OCPs and PBDEs varied widely. Pelagic and benthic food-chain components were separated by their $\delta^{13}C$ values. Significant positive correlations between $\delta^{15}N$ and PCB 153, PCB 138, *p,p′*-DDE, *oxy*-chlordane, and *trans*-nonachlordane were found only for pelagic consumers, indicating that the pelagic food chain is an important bioaccumulation pathway for selected PCB and OCP compounds. The other compounds did not show any biomagnification through benthic and pelagic food chains, suggesting the lower bioaccumulation potentials of these contaminants.

1. Introduction

Polychlorinated biphenyls (PCBs), organochlorine pesticides (OCPs), and polybrominated diphenyl ethers (PBDEs) are representative persistent organic pollutants (POPs). Their characteristics within the environment include persistence, bioaccumulation, long-range transport, toxicity, and biomagnification through food webs. Long-term exposure to PCBs, OCPs, and PBDEs elicits adverse health effects such as developmental defects, cancer, and endocrine disruption to both wildlife and humans (Kelce et al., 1995; Birnbaum and Stask, 2004; Lee et al., 2006; Ha et al., 2007). Although PCBs and OCPs have not been produced since the ban or restricted use under the Stockholm Convention in 2001, high levels of these contaminants remain in coastal environments (Sudaryanto et al., 2008; Moon et al., 2008, 2009; Won et al., 2009). The total historical consumption of OCPs, PCBs, and PBDEs in South Korea was 31,000, 9000, and 12,408 tons, respectively, in 2002 (Kim et al., 2007). The consumption of PCBs in China was reported to be around 20,000 tons in the late 1990s (Zang and Chongyano, 2000). In addition, 4.9 million tons of hexachlorocyclohexane compounds (HCHs) and 0.4 million tons of dichloro-diphenyl-trichloto-ethane and its metabolites (DDTs) were known to be produced (Zhang et al., 2002). The production of brominated flame retardants (BFRs) in China was approximately 10,000 tons in 2000, those of decabrominated diphenyl ether (deca-BDE) was 15,000 tons/yr in 2006 (Zhang et al., 2009). PBDEs are used widely as BFRs in many products such as consumer electrical goods and textiles (Watanabe and Sakai, 2003). Because of the environmental and health concerns, penta- and octa-BDE commercial mixtures have been banned in Europe and the USA since 2004. Deca-BDE technical mixtures are also banned in some European countries and in some US states (Crosse et al., 2012). A few studies on PBDEs are available from coastal and marine environments in Korea (Moon et al., 2007, 2010, 2012).

Stable isotopes provide a powerful tool for ecotoxicological studies. The nitrogen stable isotope ratio ($\delta^{15}N$) is used as an indicator of the trophic position of an animal because the $\delta^{15}N$ value increases by about 3.4‰ per trophic level as the trophic level increases in aquatic food chains (Minagawa and Wada, 1984; Michener and Schell, 1994; Hobson et al., 1995). The stable carbon isotope ratio ($\delta^{13}C$) of an animal also reflects that of its dietary source with predictable trophic enrichment (<1‰). Therefore, $\delta^{13}C$ can be used to identify the sources of carbon in marine ecosystems and to elucidate the prevalence of inshore versus offshore and/or pelagic versus benthic food sources (Hobson and Welch, 1992; Lawson and Hobson, 2000).

The Yellow Sea, which is surrounded by the Korean peninsula and China, is a shallow (<70 m in water depth) and semi-enclosed shelf. Extensive international shipping, aquaculture, and fishing

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activities are concentrated in the Yellow Sea. The industry and economy of both countries have developed rapidly over the past few decades, and thus these countries have high potential for POP contamination through riverine discharges and atmospheric transport (Lammel et al., 2007; Wang et al., 2010). Several studies have reported the contamination of sediments by POPs in the Yellow Sea (Ma et al., 2001; Yang et al., 2005; Zhang et al., 2007; Liu et al., 2008). However, measurements of the content of POPs in marine organisms such as fish and shellfish are very limited in this region (Oh et al., 2005; Kannan et al., 2010). The objectives of this study were to investigate the accumulation status of PCBs, OCPs, and PBDEs in the Yellow Sea and to use stable isotope ratios ($\delta^{13}C$ and $\delta^{15}N$) to assess the bioaccumulation potentials of these contaminants in the food web, comprising of zooplankton to cetaceans from the Yellow Sea.

2. Materials and methods

2.1. Study area

The Yellow Sea is a semi-closed continental shelf with an average depth of 44 m and surface area extent of 38×10^4 km^2 (Fig. 1). The rivers that discharge large amounts of fresh water containing suspended particles into the Yellow Sea include the Changjiang, Daliaohe, Yellow, Yalujiang, and Haihe Rivers of China, and the Han, Kum, and Yeongsan rivers of Korea (Qin et al., 1989). Various kinds of contaminants accumulated through atmospheric deposition associated with long-range transport have been reported in this region (Gao et al., 1992; Liu et al., 1998; Yeo et al., 2004). The Yellow Sea is one of the major feeding and breeding grounds of fish in northeast Asian seas. Fishery in the Yellow Sea is very intensive, with a total catch of fish reaching one million tons per year (Liu and Chen, 1998). The abundance of fish species in the Yellow Sea is low compared with other temperate waters of the same latitude. The major species for fisheries are anchovy, croaker, flatfish, herring, mackerel, and hairtail. Chinese shrimp, mantis shrimp, white-hair rough shrimp, and swimming crab are abundant invertebrates.

2.2. Sample collection and treatment

A total of 32 marine species were collected in the central part of the Yellow Sea in July 2007 (Fig. 1). Zooplankton was collected with a twin bongo net (0.6 m diameter openings, 250 μm mesh size). The net was hauled obliquely from approximately 40 m depth to the surface. The other invertebrates and fish were collected using a bottom trawl (length 41 m, width 18.8 m, 38 mm mesh, and 10 mm cod end mesh net). Organisms collected were cleaned of epibionts and identified on board. Each taxon was placed into a separate polyethylene bag, stored on ice, and transported to the land-based laboratory. All collected samples were kept frozen at −35 °C in the laboratory until subsequent treatment. After removal of the skin of the fish and cephalopods, the muscle tissues were homogenized using an ultra-disperser. The shells of bivalves, gastropods, and crustaceans were removed, and the whole soft tissues were pooled and homogenized for analysis. Muscle tissues from by-caught minke whales from the Yellow Sea in 2007 were also obtained from the Cetacean Research Institute, Korea. For stable isotope analysis, muscle tissues of all specimens were taken individually. The remaining muscle tissues of the same species were pooled and homogenized, then freeze-dried and ground to powder using a mortar and pestle for later analyses. Powdered tissue samples for $\delta^{13}C$ analysis were defatted using a mixture of methanol, chloroform, and water (2:1:0.8 by volume) according to the method of Bligh and Dyer (1959). This step avoids disrupting $\delta^{13}C$ values because of between-species differences in the concentration of isotopically lighter lipids (Focken and Becker, 1998). Zooplankton samples for $\delta^{13}C$ analysis were treated with 10% HCl to remove bicarbonate before defatting. The defatting procedure was not necessary for samples for $\delta^{15}N$ analysis. Animal tissue samples for isotope analysis to be processed were dried in an

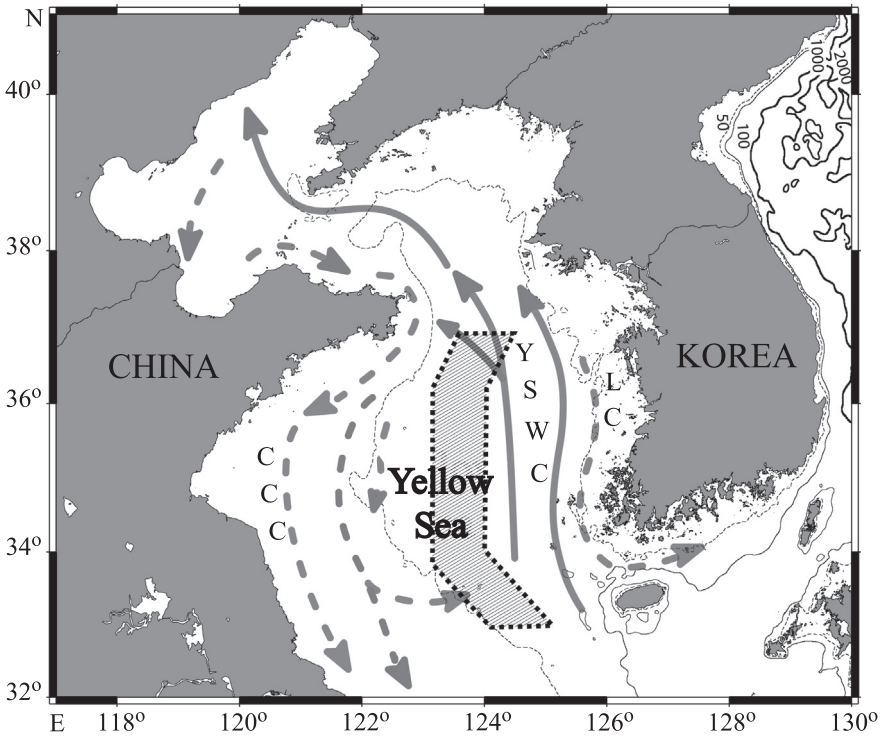

Fig. 1. Map showing the study area in the central region of the Yellow Sea. CCC, China Coastal Current; YSWC, Yellow Sea Warm Current; LC, Littoral Current.

oven at 60 °C and kept frozen (−70 °C) in the laboratory until subsequent treatment.

2.3. Sample preparation

The concentrations of 21 PCB congeners (PCBs 18, 28, 29, 44, 52, 87, 101, 105, 110, 118, 128, 138, 153, 170, 180, 187, 194, 195, 200, 205, and 206), 24 PBDE congeners (BDEs 17, 28, 47, 49, 66, 71, 77, 85, 99, 100, 119, 126, 138, 153, 154, 156, 183, 184, 191, 196, 197, 206, 207, and 209), and 15 OCP compounds were measured in all marine organism samples. DDTs included *p,p′*-DDE, *o,p′*-DDE, *p,p′*-DDE, *o,p′*-DDE, *p,p′*-DDT, and *o,p′*-DDT; chlordanes (CHLs) included *oxy*-CHL, *trans*-CHL, *cis*-CHL, *trans*-nonaCHL, and *cis*-nonaCHL; and HCHs included α-, β-, and γ-HCH. Hexachlorobenzene (HCB) was also analyzed.

The details of the experimental procedures to analyze PCBs, OCPs, and PBDEs in marine organisms have been reported elsewhere (Moon et al., 2007, 2009). In brief, about 20 g of each sample was homogenized with anhydrous Na_2SO_4 and extracted using a Soxhlet extractor for 17 h with a 3:1 mixture of dichloromethane (ultra-residue analysis, J.T. Baker, Phillipsburg, NJ, USA) and hexane (ultra-residue analysis, J.T. Baker). Before the extraction, surrogates of PCBs 103, 198, and 209 were spiked into the samples. The extract was concentrated to 11 mL using a rotary evaporator. An aliquot of extracted samples was subsampled for gravimetric measurement of lipid content. Each 5 ng of the internal standards $^{13}C_{12}$-labeled PBDE congeners (MBDE-MXE; Wellington Laboratories, Guelph, ON, Canada), $^{13}C_{12}$-labeled PCB congeners (MBP-MXE; Wellington Laboratories), and OCP congeners (EC-5349; Cambridge Isotope Laboratories, Andover, MA, USA) were spiked into the remaining extracts (10 mL). Lipid in the sample extract was removed by gel permeation chromatography using a Bio-beads S-X3 (Bio-Rad Laboratories, Hercules, CA, USA) packed glass column (380 mm × 22 mm inner diameter) with a successive cartridge packed with 0.5 g of silica gel (neutral, 70−230 mesh, Merck, Darmstadt, Germany). Each extract was cleaned up with a multilayer silica gel column containing 10% $AgNO_3$ silica gel, 22% H_2SO_4 silica gel, 44% H_2SO_4 silica gel, and 2% KOH silica gel (GL Sciences, Tokyo, Japan). The eluents were concentrated to approximately 1 mL and evaporated at room temperature to 50–100 μL. Five nanograms of $^{13}C_{12}$-labeled PCB recovery standard (EC 9605; Wellington Laboratories) was added before instrumental analysis. The residues were transferred to 50 μL of *n*-nonane (pesticide analysis grade, Fluka, St. Gallen, Switzerland) for instrumental analysis.

2.4. High-resolution gas chromatograph/high-resolution mass spectrometer (HRMS) analysis

Identification and quantification of PCBs, OCPs, and PBDEs were performed using a high-resolution gas chromatograph interfaced with a high-resolution mass spectrometer (HRMS; JMS800D, JEOL, Tokyo, Japan). The details of the instrumental analyses have been presented elsewhere (Moon et al., 2007, 2009). PCBs, OCPs, and PBDEs were quantified using the isotope dilution method, based on the relative response factors of individual compounds. The HRMS was operated in electron ionization mode, and ions were monitored by selected ion monitoring. A DB5-MS gas chromatograph column (30 m length, 0.25 mm inner diameter, 0.25 μm film thickness; J&W Scientific, Palo Alto, CA, USA) was used to separate PCBs and OCPs. PBDE congeners, ranging from tri- to hepta-BDEs and from octa- to deca-BDEs, were quantified separately using a DB5-MS column (15 m length, 0.25 mm inner diameter, 0.1 μm film thickness; J&W Scientific).

Solvents injected before and after the injection of standards showed negligible contamination or carryover. Procedural blanks ($n = 7$), which were processed using the same procedure as the real samples, did not contain quantifiable amounts of target compounds except for BDE 209, which was present at ~0.1 ng g^{-1}. The recovery of surrogates (PCBs 103, 198, and 209) spiked before the extraction was 73 ± 8.9% (average ± SD). The calculated limits of detection (signal-to-noise ratio = 3) were 1 ng g^{-1} for OCPs, 0.04−0.08 pg g^{-1} for PCBs, and 0.1−0.5 pg g^{-1} for tri- to octa-BDEs. To quantify deca-BDE concentration, the average levels measured in the procedural blanks were subtracted from the concentrations detected in the samples. To assess the quality of the experimental procedures and instrumental conditions, the standard mussel (*Mytilus edulis*) reference material (SRM 2977; NIST, Gaithersburg, MD, USA) was analyzed for PCBs, OCPs, and PBDEs. The recovery ($n = 4$) ranged from 64% to 87% for PCBs, 86% to 116% for OCPs, and 87% to 107% for PBDEs.

2.5. Stable isotope analysis and trophic magnification factors (TMFs)

Carbon and nitrogen isotope ratios were determined using a continuous-flow isotope-ratio mass spectrometer aligned with an elemental analyzer. Dried subsamples (0.5–1.5 mg) were weighed in tin capsules (EuroVector, 6 × 4 mm). The samples wrapped in tin capsules were oxidized at 1030 °C in an elemental analyzer (EuroVector 3000 series, Milan, Italy), and the resultant CO_2 and N_2 gases were analyzed for stable isotope ratios with a continuous-flow isotope-ratio mass spectrometer (CF-IRMS, Micromass IsoPrime, Manchester, UK). Stable isotope ratios are expressed as the relative parts per thousand (‰) differences between the samples and standard reference materials (Pee Dee Belemnite for carbon and atmospheric N_2 for nitrogen) using the following equation:

$$\delta X = [(R_{sample}/R_{standard}) - 1] \times 10^3,$$

where X is ^{13}C or ^{15}N, and R is the corresponding ratio of ^{13}C:^{12}C or ^{15}N:^{14}N. IAEA CH-6 (sucrose, $\delta^{13}C = -10.1 \pm 0.2$‰) and IAEA-N1 (ammonium sulfate, $\delta^{15}N = 2.8 \pm 0.3$‰) were used as reference materials. Measurement precision for 20 repeated analyses was ~0.1‰ and 0.2‰ for $\delta^{13}C$ and $\delta^{15}N$, respectively.

The trophic level (TL) of consumer species was calculated by the following formula:

$$TL_{consumer} = ((\delta^{15}N_{consumer} - \delta^{15}N_{zooplankton})/3.4) + 2.$$

Zooplankton was assumed to be a primary consumer (defined as inhabiting TP 2) as a trophic baseline. Because trophic enrichment in ^{15}N is more variable for primary consumers than for carnivores, using primary consumers as a baseline might reduce error in the calculation of trophic level (Post et al., 2000; Vander Zanden and Rasmussen, 2001).

TMFs have been used recently as a more reliable method to assess bioaccumulation potentials of contaminants, especially biomagnification in the food web (Borgå et al., 2012; Hallanger et al., 2011) because TMFs can be investigated independently of the initial exposure level (Broman et al., 1992). In our study, TMFs were estimated as the antilog of the regression slope (b) with base 10 (TMF = 10^b) of the linear regression between the log concentration of the target contaminants (lipid wt) and TL of the sample using the following formula as suggested by Borgå et al. (2012) and Hallanger et al. (2011):

$$\log[POPs] = a + bTL.$$

2.6. Statistics

The concentrations of PCBs, OCPs, and PBDEs were adjusted to the lipid content contained in the samples for each organism. Spearman's rank correlation analysis was performed to investigate

relationships among target contaminants, and between contaminants and $\delta^{15}N$ in marine organisms. The commercially available SPSS package (IBM Corp., Armonk, NY, USA) was used for the statistical analysis.

3. Results and discussion

3.1. Contamination status of PCBs, OCPs, and PBDEs

The biological information on 32 specimens collected from the Yellow Sea is summarized in Table 1. The lipid content on a wet weight basis ranged from 1.3% (*Neptunea cumingi*) to 21% (*Conger myriaster*), with a narrow range within taxonomic groups. All chlorinated and brominated contaminants including DDTs, PCBs, CHLs, HCHs, HCB, and PBDEs were detected in all 32 marine species analyzed (Table 2). The concentration of DDTs (6.9–357 ng g^{-1} lipid weight, mean 139 ng g^{-1} in all the samples except for minke whales) was the highest among the POPs analyzed. The concentrations of DDTs were one to two orders of magnitude higher than those of other contaminants. Because large amounts of DDT have been used for agricultural purposes in China, the Yangtze riverine discharges into the Yellow Sea could be a major source of DDT contamination in marine biota in this region (Zhang et al., 2007). The concentrations of PCBs, HCHs, HCB, and CHLs were in the range of 1.5–32 (mean 11) ng g^{-1} lipid weight, 1.0–40 (mean 7.5) ng g^{-1} lipid weight, 0.6–9.9 (mean 4.6) ng g^{-1} lipid weight, and 0.4–8.7 (mean 2.7) ng g^{-1} lipid weight, respectively. The concentrations of PBDEs ranged from 0.5 to 63 (mean 8.9) ng g^{-1} lipid weight, which were similar to those of HCHs. The contamination patterns of these contaminants were similar to those found in previous studies of marine organisms from Korean and Chinese coastal environments (Liu et al., 2008; Moon et al., 2009, 2010). The concentrations of deca-BDE were in the range of not detectable to 20 ng g^{-1} lipid weight. These values were several times higher than the concentrations of tri- to nonaBDEs. The concentrations of PBDEs in our study were lower than those found in commercial fish from China (0.8–69 ng g^{-1} lipid weight, Meng et al., 2007) and marine organisms from Bohai Bay, China (0.15–33 ng g^{-1} lipid weight, Wan et al., 2008).

The concentrations of PCBs and OCPs measured in our study were compared with those reported for the coastal/offshore waters of Northeast Pacific regions (Table 3). The overall contamination status of PCBs and OCPs in our study was similar to that found in squid from the Yellow Sea (Won et al., 2009). However, the concentrations of PCBs and OCPs in fish and shellfish from Chinese and Japanese waters were one or two orders of magnitude higher than those measured in our study (Jin et al., 2008; Takahashi et al., 2010), probably because of the use of large amounts of OCPs in China. Indeed, 0.4 million tons of DDTs and their metabolites have

Table 1
Sample details of various marine organisms from the Yellow Sea.

No.	Species	Common name	n	Body size (cm)	Moisture (%)	Lipid (%)
1	Zooplankton (mixed)		1	–	–	17
Fish						
2	*Pholis fangi*	White blenny	68	15 ± 0.9	64	4.1
3	*Psenopsis anomala*	Pacific rudderfish	23	15 ± 0.6	67	3.7
4	*Apogon lineatus*	Verticalstriped cardinalfish	37	8.7 ± 0.9	68	4.6
5	*Paralichthys olivaceus*	Oliver flounder	1	38	72	2.0
6	*Hexagrammos otakii*	Greenling	19	22 ± 1.7	69	4.3
7	*Okamejei kenojei*	Ocellate spot skate	6	33 ± 3.1	66	1.7
8	*Conger myriaster*	Conger eel	11	34 ± 5.9	58	21
9	*Trichiurus lepturus*	Largehead hairtail	21	54 ± 7.4	67	4.4
10	*Scomber japonicas*	Chub mackerel	1	26	55	8.3
11	*Trachurus japonicas*	Jack mackerel	9	14 ± 1.1	59	5.3
12	*Pleuronichthys cornutus*	Finespotted flounder	41	13 ± 1.0	66	1.5
13	*Larimichthys polyactis*	Small yellow croaker	28	16 ± 1.4	54	10
14	*Chelidonichthys spinosus*	Bluefin searobin	15	17 ± 4.1	58	2.8
Cephalopod						
15	*Euprymna morsei*	Mimika bobtail	37	3.5 ± 0.8	63	4.1
16	*Loligo japonica*	Japanese squid	10	19 ± 1.4	75	2.9
17	*Octopus variabilis*	Whiparm octopus	4	63 ± 22	78	1.5
Bivalve						
18	*Scapharca broughtonii*	Broughton's ribbed ark	2	6.5 ± 0.6	48	1.7
19	*Panopea japonica*	Japanese geoduck	1	12	69	1.9
Gastropod						
20	*Neptunea cumingi*	Arthritic neptune	17	11 ± 11	69	1.3
21	*Siphonalia fusoides*	Fusiform whelk	92	5.2 ± 0.5	74	1.4
22	*Psephaea kaneko*	Kaneko volute	3	18 ± 1.2	73	1.4
Crustacea						
23	*Oratosquilla oratoria*	Mantis shrimp	17	11 ± 1.1	67	2.9
24	*Palaemon gravieri*	Chinese ditch prawn	171	5.0 ± 0.4	68	1.5
25	*Crangon hakodatei*	Hakodate sand shrimp	205	8.3 ± 0.8	76	1.7
26	*Solenocera melantho*	Big head shrimp	75	9.5 ± 0.7	70	1.7
27	*Pagurus ochotensis*	Alaskan hermit	61	10 ± 0.6	58	1.3
28	*Oregonia gracilis*	Decorator crab	23	3.9 ± 0.6	68	5.9
29	*Charybdis bimaculata*	Two-spot swimming crab	78	2.9 ± 0.3	67	4.3
30	*Ovalipes punctatus*	Sand crab	15	3.7 ± 1.4	66	6.0
31	*Portunus trituberculatus*	Swimming crab	1	20	66	3.3
Cetacean						
32	*Balaenoptera acutorostrata*	Minke whale (male)	6	7333 ± 234	–	6.6 ± 5.2
32-1	*Balaenoptera acutorostrata*	Minke whale (female)	4	6200 ± 1230	43 ± 2.7	9.7 ± 0.6

–: Not measured.

Table 2
Concentrations of organohalogen compounds (ng g^{-1} lipid weight), $\delta^{13}C$ and $\delta^{15}N$ values in marine organisms from the Yellow Sea.

No.	Common name	∑DDT	∑PCB	∑HCH	HCB	∑CHL	∑PBDE	$\delta^{15}N$	$\delta^{13}C$
1	Zooplankton	39	1.5	1.0	0.7	0.5	5.5	7.5	−19.1
Fish									
2	White blenny	308	14	8.7	9.9	3.6	12	11.8	−18.4
3	Pacific rudderfish	46	3.8	4.5	2.3	6.9	1.7	12.4	−13.6
4	Verticalstriped cardinalfish	144	12	7.9	6.4	4.6	5.7	11.4	−16.6
5	Oliver flounder	197	11	5.5	4.3	2.1	3.9	12.1	−14.3
6	Greenling	357	26	15	9.3	3.8	12	12.4	−18.0
7	Ocellate spot skate	26	2.8	3.7	1.9	0.7	1.4	11.5	−15.7
8	Conger eel	292	11	8.8	5.4	2.9	5.1	13.3	−17.2
9	Largehead hairtail	214	8.4	10	4.5	2.9	7.5	11.9	−15.6
10	Chub mackerel	336	8.8	18	6.4	2.9	6.2	9.5	−18.5
11	Jack mackerel	60	3.1	5.5	3.0	2.5	2.2	12.2	−14.7
12	Finespotted flounder	122	7.5	13	6.6	1.1	4.7	10.5	−16.2
13	Small yellow croaker	247	11	7.5	6.5	3.9	11	10.9	−17.1
14	Bluefin searobin	130	8.6	5.0	4.5	2.1	6.6	12.1	−15.3
Cephalopod									
15	Mimika bobtail	177	6.3	2.9	5.2	4.4	10	10.5	−16.6
16	Japanese squid	75	7.1	1.5	2.7	1.5	9.4	12.9	−13.2
17	Whiparm octopus	63	9.3	1.5	4.0	0.8	8.2	11.4	−15.9
Bivalve									
18	Broughton's ribbed ark	48	8.9	4.0	3.1	0.9	3.2	8.3	−16.2
19	Japanese geoduck	108	7.9	40	5.1	1.6	5.9	8.6	−17.1
Gastropod									
20	Arthritic neptune	158	12	2.1	2.6	2.1	5.5	11.4	−14.3
21	Fusiform whelk	70	15	3.1	6.9	1.2	2.3	12.1	−14.4
22	Kaneko volute	42	9.8	2.6	1.8	1.5	2.5	12.8	−14.8
Crustacea									
23	Mantis shrimp	61	16	5.2	0.6	3.0	6.1	12.5	−14.3
24	Chinese ditch prawn	47	5.9	3.8	4.5	1.6	9.3	10.7	−14.6
25	Hakodate sand shrimp	69	5.5	4.1	1.7	0.9	16	10.5	−17.2
26	Big head shrimp	6.9	5.3	1.5	2.5	0.4	5.4	11.3	−16.2
27	Alaskan hermit	105	14	17	3.3	3.7	14	11.2	−16.8
28	Decorator crab	314	32	13	7.5	4.0	5.2	11.4	−17.5
29	Two-spot swimming crab	147	26	2.3	8.0	4.3	6.2	10.4	−16.9
30	Sand crab	121	17	9.5	7.1	4.0	15	11.3	−16.0
31	Swimming crab	172	9.1	3.4	3.4	8.7	2.2	11.4	−16.2
Cetacean									
32	Minke whale (male)	1581 ± 1038	690 ± 398	189 ± 279	39 ± 20	80 ± 46	115 ± 62	10.9	−17.9
32-1	Minke whale (female)	202 ± 129	20 ± 13	48 ± 50	5.4 ± 4.4	5.3 ± 4.3	9.4 ± 0.9	10.5	−16.5

Table 3
Comparison of concentrations (range and mean, ng g^{-1} lipid wt) of PCBs and OCPs in the marine biota from northeast Asian regions.

Location	Species	Year	DDTs	PCBs	HCHs	HCB	CHLs	Reference
Northeast coast of China	Bivalves	2005	2240–33,086 (769)	53–314 (157)	33–296 (151)		nd-24 (10)	Jin et al. (2008)
Yellow Sea (Korea)	Squid	2006	118–698 (326)	70–275 (131)	3.0–11 (7.0)	5.0–8.0 (6.0)	4.0–22 (8.0)	Won et al. (2009)
Western north pacific (Japan)	Deep-sea fish	2005	14–830 (110)	nd-2200 (150)	nd-150 (25)	nd-100 (21)	3.9–640 (54)	Takahashi et al. (2010)
Yellow Sea	Marine biota	2007	6.9–357 (139)	1.5–32 (11)	1.0–40 (7.5)	0.6–9.9 (4.6)	0.4–8.7 (2.7)	This study

been produced in China (Zhang et al., 2002), and technical CHLs are still being used for termite extermination in China (Xu et al., 2004). HCHs and HCB are also produced and used in China, and consequently intense pollution by these chemicals is expected in adjacent environments (Wu et al., 1997a).

Pearson correlation coefficients showed significant relationships between DDTs, PCBs, CHLs, HCHs, HCB, and PBDEs ($r = 0.924–0.980$, $p < 0.001$). This finding suggests that these contaminants have similar sources and/or bioaccumulation behavior in the marine food web.

In this study, significantly higher levels of contaminants were found in male minke whales (*Balaenoptera acutorostrata*) than in female minke whales, indicating a sex difference in the concentrations of POPs. Previous studies confirmed that maternal transfer and lactation are the major excretion processes for PCBs, OCPs, and PBDEs in female mammals (Aguilar et al., 1999; Moon et al., 2010; Park et al., 2010).

3.2. Accumulation features of PCBs, OCPs, and PBDEs

The accumulation profiles of PCBs, OCPs, and PBDEs in marine species from the Yellow Sea are shown in Fig. 2. The accumulation patterns of PCBs were relatively uniform in the species examined, suggesting that PCBs have a greater stability for metabolic transformation (Moon et al., 2010). The predominant congeners of PCBs were PCBs 153, 138, and 118, which collectively accounted for 44%

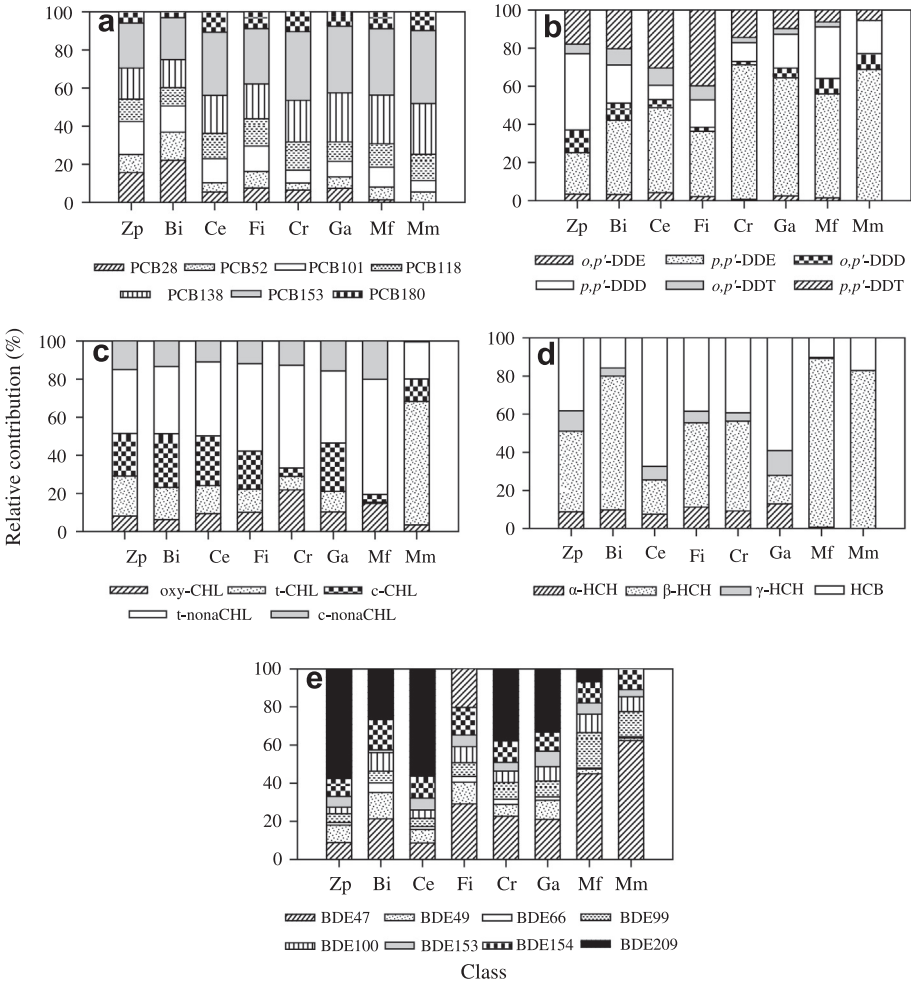

Fig. 2. Accumulation profiles of PCBs, OCPs, and PBDEs in marine species from the Yellow Sea. Zp, zooplankton; Bi, bivalve; Ce, cephalopod; Fi, fish; Cr, crustacean; Ga, gastropod; Mf, female minke whale; Mm, male minke whale.

of the total PCB concentrations; these values are consistent with data in previous studies (Moon et al., 2009; Takahashi et al., 2010). A number of studies have reported that the bioaccumulation potentials of organochlorine (OC) compounds are associated with the octanol–water partition coefficient ($\log K_{ow}$) (Fisk et al., 2001; Moisey et al., 2001; Goerke et al., 2004; Nfon et al., 2008). Nfon et al. (2008) reported that the greatest accumulation values for OCs are found in the $\log K_{ow}$ range of 6.5 to 7.0 and decrease when > 7.5 of $\log K_{ow}$. The $\log K_{ow}$ values of the tri- to penta-CBs range from 5.5 to 6.5, and the hexa- to deca-CBs range from 6.7 to 8.26 (Mackey et al., 1992). The accumulation patterns of PCBs found in our study were characterized by the dominance of hexa- and hepta-CBs, which ranged from 6.7 to 7.6 of $\log K_{ow}$, except for zooplankton and bivalves. Our observation indicates that the accumulation features of PCB in marine species from the Yellow Sea are governed by the physicochemical properties (K_{ow}) of these contaminants.

The contributions of individual OCP compounds varied widely between marine species, indicating a lower stability compared with PCBs. For DDTs, the major compounds were *p*,*p*′-DDE, *p*,*p*′-DDT, and *p*,*p*′-DDD, which collectively accounted for 85% of DDTs (Fig. 2b). *p*,*p*′-DDE and *p*,*p*′-DDD are metabolites of DDT in the environment and biota. Among CHL and HCH isomers, *trans*-nonaCHL and β-HCH were the major components (Fig. 2c and d). The predominance of these contaminants in marine species is associated with metabolism. In general, the compounds with a $\log K_{ow} < 5.5$ (HCHs, CHLs, and HCB) have lower capacities for bioaccumulation in marine organisms (Mackey et al., 1999). However, because of their limited metabolism, β-HCH and *oxy*-, *trans*-, and *cis*-nonaCHL have higher bioaccumulation potentials relative to other isomers of HCHs and CHLs (Wu et al., 1997b; Willett et al., 1998). Similar results have been found in other studies (Ruus et al., 1999; Fisk et al., 2001; Hop et al., 2002; Ikemoto et al., 2008).

The accumulation patterns of PBDE congeners differed between marine species because of differing stabilities of PBDE congeners (Fig. 2e). In particular, fish and male minke whales did not contain BDE 209 because of the lower bioaccumulation potentials for these species. However, other species such as bivalves and crustaceans showed a predominance of BDE 209 in the total PBDE concentration. Mizukawa et al. (2009) reported that BDE 209 may not permeate through biological membranes because of its large molecular size (MW = 959). The high proportion of BDE 209 in some marine species may be associated with the high consumption of deca-BDE in coastal zones of Korea. Korea accounts for 50% of total deca-BDE use in Asia because of the rapid growth of its electronic industries (Moon et al., 2007, 2010). In addition, the greatest contamination by PBDEs has been reported in Chinese coastal waters such as the Pearl River Delta and Bo Sea (Mai et al., 2005; Wang et al., 2009). Therefore, the predominance of deca-BDE in various species reported here seems to relate to the local and ongoing sources of PBDE products in the Yellow Sea coastal region.

3.3. Bioaccumulation potentials of PCBs, OCPs, and PBDEs

The $\delta^{13}C$ and $\delta^{15}N$ values ranged from −19.1‰ in zooplankton to −13.2‰ in Japanese squid (*Loligo japonica*) and 7.5‰ in zooplankton to 13.3‰ in conger eel (*C. myriaster*), respectively (Table 2). Zooplankton, which constitutes an important prey item of higher-trophic organisms in marine ecosystems, had the lowest $\delta^{13}C$ and $\delta^{15}N$ values of all the specimens analyzed. Bivalves, which are characterized as suspension feeders that consume plankton and other organic detritus, had lower $\delta^{15}N$ values (8.3‰ in *Scapharca broughtonii* and 8.6‰ in *Panopea japonica*) compared with other species.

The dual-isotope plot of $\delta^{13}C$ and $\delta^{15}N$ of all consumers analyzed differentiated two trophic pathways (Fig. 3). Based on the $\delta^{13}C$ values, two groups that may use different carbon sources were identified. One group, whose $\delta^{13}C$ values ranged from −19.1 to −18.0‰, comprised pelagic consumers including zooplankton, white blenny (*Pholis fangi*), greenling (*Hexagrammos otakii*), and chub mackerel (*Scomber japonicus*). The other group, with $\delta^{13}C$ values of −17.0 to −13.2‰, comprised benthic consumers including bivalve, cephalopod, crustacean, gastropod, and the other fish species. It seems to be plausible to divide the organisms into two groups (i.e., pelagic and benthic food-chain components) based on their $\delta^{13}C$ values. Separation by $\delta^{13}C$ values between pelagic and benthic feeding groups has been observed elsewhere (France, 1995; Yoshii et al., 1999; Hobson et al., 2002; Mincks et al., 2008). For example, the $\delta^{13}C$ values of two bivalves (*S. broughtonii* and *P. japonica*), which constitute the trophic baseline of the benthic food web, were 2–3‰ higher than the mean value of −19.1‰ in zooplankton. Moreover, $\delta^{13}C$ values of Japanese squid (*L. japonica*) and Pacific rudderfish (*Psenopsis anomala*) were up to 5‰ higher than those of pelagic feeding fish, white blenny, and greenling. The ^{13}C-enrichment in the benthic food chain may be explained by substantial reworking of sedimentary organic matter before uptake by benthic fauna (Mincks et al., 2008). The $\delta^{13}C$ values in particulate organic matter at the sediment–water boundary can be shifted by 1–4‰ because of the intensity and/or mode of benthic remineralization (Fischer, 1991). Furthermore, benthic, detritus-based food webs display 4–5‰ greater enrichment of $\delta^{13}C$ relative to particulate organic matter compared with pelagic consumers (Hobson et al., 1995; Nyssen et al., 2002; Mincks et al., 2008). These authors presumed that such a great ^{13}C-enrichment in the benthic food chain can be explained by the assimilation of microbial and/or meiofaunal biomass and the strong carbon fractionation effects of bacterial metabolism.

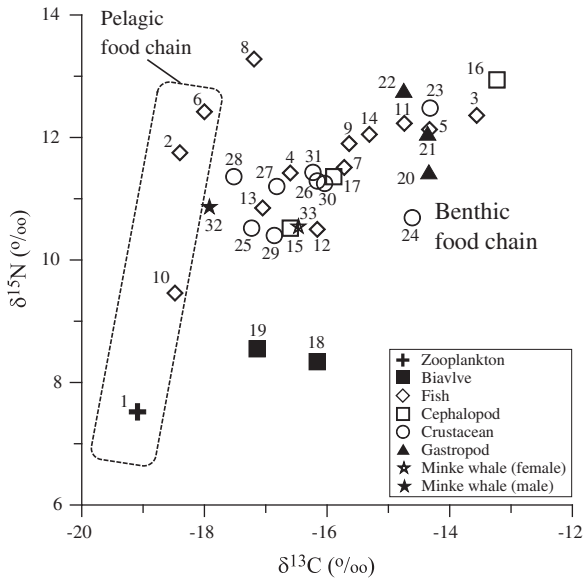

Fig. 3. Dual plot of $\delta^{13}C$ and $\delta^{15}N$ measured for zooplankton, bivalve, fish, cephalopod, crustacean, gastropod, and minke whale from the Yellow Sea. The dotted box indicates the pelagic food chain, assuming the general trophic fractionation coefficients of +1‰ for $\delta^{13}C$ and +3.4‰ for $\delta^{15}N$ at each trophic transfer (Minagawa and Wada, 1984; Michener and Schell, 1994). 1 zooplankton; **Fish**: 2 *Pholis fangi*, 3 *Psenopsis anomala*, 4 *Apogon lineatus*, 5 *Paralichthys olivaceus*, 6 *Hexagrammos otakii*, 7 *Okamejei kenojei*, 8 *Conger myriaster*, 9 *Trichiurus lepturus*, 10 *Scomber japonicas*, 11 *Trachurus japonicas*, 12 *Pleuronichthys cornutus*, 13 *Larimichthys polyactis*, 14 *Chelidonichthys spinosus*; **Cephalopod**: 15 *Euprymna morsei*, 16 *Loligo japonica*, 17 *Octopus variabilis*; **Bivalve**: 18 *Scapharca broughtonii*, 19 *Panopea japonica*; Gastropod: 20 *Neptunea cumingi*, 21 *Siphonalia fusoides*, 22 *Psephaea kaneko*; **Crustacean**: 23 *Oratosquilla oratoria*, 24 *Palaemon gravieri*, 25 *Crangon hakodatei*, 26 *Solenocera melantho*, 27 *Pagurus ochotensis*, 28 *Oregonia gracilis*, 29 *Charybdis bimaculata*, 30 *Ovalipes punctatus*, 31 *Portunus trituberculatus*; **Minke whale**: 32 male *Balaenoptera acutorostrata*, 33 female *Balaenoptera acutorostrata*.

As mentioned earlier, the meiofaunal $\delta^{15}N$ of an animal increases by 3–5‰ at each trophic transfer, with an average of 3.4‰ for a wide variety of taxa (Minagawa and Wada, 1984; Michener and Schell, 1994; Hobson et al., 1995). Because of this relatively large trophic fractionation, the measurement of $\delta^{15}N$ has been used as a numerical representative of trophic position (Post, 2002). Since the $\delta^{15}N$ value of an animal reflects assimilated dietary integration over time, $\delta^{15}N$ can be a sensitive trophic descriptor and thus a good predictor of contaminant biomagnification (Cabana and Rasmussen, 1994; Atwell et al., 1998; Hobson et al., 2002). These authors suggested that the bioaccumulation potentials and trophic transfer of organic contaminants through aquatic food webs can be examined by the relationship between $\delta^{15}N$ and contaminant levels in the tissues of organisms.

Pelagic and benthic consumer groups showed different biomagnification patterns of specific compounds (or congeners) of PCBs, OCPs, and PBDEs. In the pelagic feeding group (zooplankton, *P. fangi*, *H. otakii*, and *S. japonicas*), the concentrations of the major congeners such as PCBs 153, 138, *p,p′*-DDE, *oxy*-CHL, and *trans*-nonaCHL were significantly higher with increasing $\delta^{15}N$ values ($p < 0.01$, Spearman's rank correlation test, Fig. 4a). These compounds are generally known to have a high food-chain magnification factor within the $\log K_{ow}$ range of 5.0 to 7.0 (Goerke et al., 2004; Nfon et al., 2008). The remaining compounds, including all the PBDE congeners, did not display any relationships with $\delta^{15}N$ (data not shown). In the benthic feeding group, there were no significant relationships between any POP and the $\delta^{15}N$ value ($p > 0.05$, Fig. 4b). This result can likely be attributed to the narrower $\delta^{15}N$ range of 10.4‰ (*Charybdis bimaculata*) to 12.9‰ (*L. japonica*) in most of the consumers of this group compared with the pelagic food-chain counterparts.

PCBs 153 and 138, *p,p′*-DDE, *oxy*-CHL, and *trans*-nonaCHL in pelagic species had significantly higher TMF values (PCB 153, 8.3; PCB 138, 8.0; *p,p′*-DDE, 5.6; *oxy*-CHL, 5.6; *trans*-nonaCHL, 4.6) compared with benthic species. Despite TMF values > 1 for several congeners (PCB 180, 1.23; *p,p′*-DDT, 1.08; BDE 209, 1.11), benthic components had TMF values < 1 for most target compounds or congeners. To understand better the status of biomagnification in the study area, the TMF values of PCBs 153 and 138, *p,p′*-DDE, *oxy*-CHL, and *trans*-nonaCHL in the pelagic food web from the Yellow Sea were compared with those reported for different food webs (Table 4). The TMF values estimated in our study, except for *trans*-nonaCHL, were similar to those found in the Northwater polynya food web (Fisk et al., 2001). Our TMF values were significantly lower than those found in the Arctic marine food web but were higher than those found in the Barents Sea food web. Such a narrow $\delta^{15}N$ range in benthic consumer group indicates that they occupy nearly the same ecological niche and thereby have similar levels of biomagnification for the contaminants analyzed.

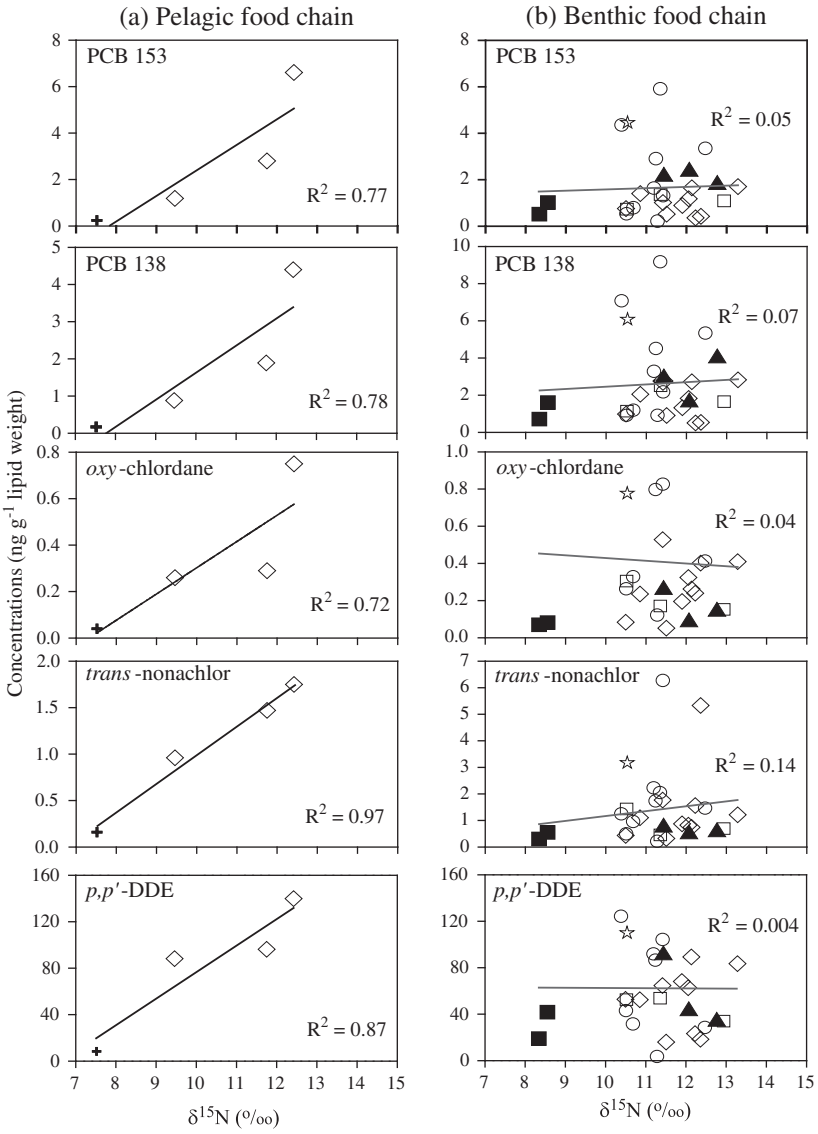

Fig. 4. Relationships between $\delta^{15}N$ and the concentrations of PCB 153, PCB 138, *p,p′*-DDE, *oxy*-CHL, and *trans*-nonaCHL (ng g^{-1} lipid weight) in (a) pelagic and (b) benthic consumers from the Yellow Sea.

Table 4
Comparison of trophic magnification factors (TMFs) between the Yellow Sea and other locations.

Location	PCB153	PCB138	*oxy*-CHL	*tran*-nonaCHL	*p,p′*-DDE	Reference
Arctic sea	32	50	23	4.5	11	Hallanger et al. (2011)
Northwater Polynya	9.7	8.8	6.5	5.5	14	Fisk et al. (2001)
Barents Sea	4.1	3.7	4.4	5.0	3.7	Hop et al. (2002)
Yellow Sea	8.3	8.0	5.6	4.6	5.6	This study

The limited food-chain biomagnification has been previously observed in other studies and might reflect the metabolic capability of higher-trophic invertebrates and crustaceans, which constitute important components of benthic assemblages (Kidd et al., 2001; Nfon et al., 2008; references therein). Metabolic characteristics of benthic prey organisms could explain the low OC concentrations in fish that have a strong trophic link to the benthic food chain (Table 2). The concentrations of OCs in higher-trophic-level consumers may also be explained by processes at the trophic base (i.e., algal growth conditions) of the food web, particle dilution of the contaminants, and growth rates of the consumers (Kidd et al., 2001). However, more precise information on the effects of these factors on the biomagnification of contaminants is lacking at present.

The $\delta^{15}N$ values (10.9 ± 0.4 for males and 10.5 ± 0.5 for females) of minke whale (*B. acutorostrata*) were similar to or lower than those of fish and invertebrates. The main prey items of *B. acutorostrata* are small invertebrates and fish such as shrimp and anchovy, as characterized for the baleen whale (Stewart and Leatherwood, 1985). However, the $\delta^{15}N$ values of many replicates are similar to or lower than those of shrimp, prawns, and small fishes in the present study. The Yellow Sea is an important habitat of minke whales in western North Pacific (the East/Japan Sea–Yellow Sea–East China Sea, Tamura and Fufise, 2002). In the Yellow Sea, it is known

that the female minke whales move to the northern part of the Yellow Sea in October and stay there until next July, and they leave the Yellow Sea with pups after delivery. Most of adults of male minke whale do not move to northern part of the Yellow Sea in the same season (Peilie, 1985). Considering the trophic fractionation effect, their low $\delta^{15}N$ values suggest limited foraging activity in this study area, further indicating that they have true feeding grounds in other locations. Their active migration may complicate use of the $\delta^{15}N$ approach to identify their trophic position. Despite their relatively low $\delta^{15}N$ values, *B. acutorostrata* had higher concentrations of all contaminants than those found in other species in the present investigation (Table 2), possibly because of their huge amount of prey consumption and relatively low metabolic activities (Tanabe, 2002; Tamura and Fufise, 2002; Moon et al., 2010).

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