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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

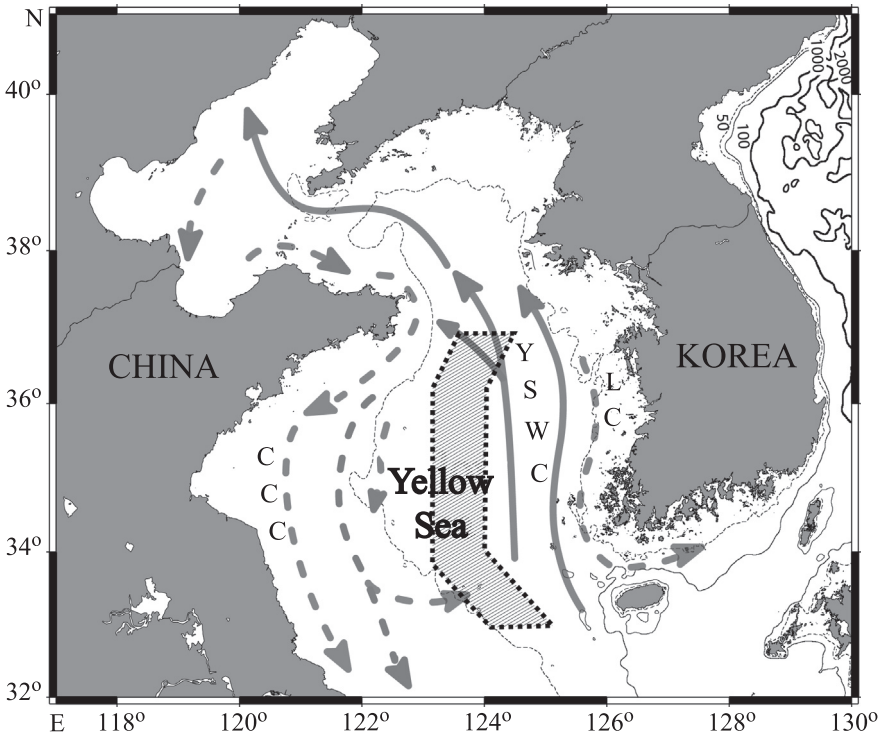

Fig. 1. Map showing the study area in the central region of the Yellow Sea. CCC, China Coastal Current; YSWC, Yellow Sea Warm Current; LC, Littoral Current.