

Chlorinated and brominated organic compounds in cetaceans from Korean coastal waters

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ABSTRACT

The livers and blubbers of minke whale (*Balaenoptera acutorostrata*) and common dolphin (*Delphinus* spp.) entangled in fishing gear were collected along the Korean coasts in 2006, to investigate levels and accumulation profiles of polychlorinated biphenyls (PCBs), organochlorine pesticides (OCPs) and polybrominated diphenyl ethers (PBDEs). Among the OCs analyzed, the levels of PCBs and DDTs were predominant in all of the samples and their concentrations were higher than one or two orders of magnitude of the concentrations of CHLs, HCHs and HCB. The concentrations of PBDEs were lower than those of PCBs and DDTs. The overall contamination of PCBs, OCPs and PBDEs in cetaceans from Korean coastal waters was relatively lower or similar to the levels of these contaminants measured from other countries. The concentrations of PCBs, DDTs, CHLs and PBDEs in the livers and blubbers of common dolphin were significantly ($p < 0.05$) higher than those measured in minke whale, while CHLs and HCHs did not show significant differences in the livers and blubbers. The predominant compounds of PCBs, OCPs and PBDEs in both cetacean species and tissue types were PCB 153, *p,p'*-DDE and BDE 47, respectively. The concentrations of PCBs and DDTs in the present study can have adverse effects such as low sex hormone and suppression of the immune system. Therefore, continuous monitoring and ecotoxicological studies are needed to understand the status of organic contamination as well as actual health effects of anthropogenic chemicals on this population of cetaceans in Korea.

KEYWORDS: COMMON DOLPHIN; MINKE WHALE; PCBs; OCPs; PBDEs; KOREAN COASTAL WATERS

INTRODUCTION

Polychlorinated biphenyls (PCBs) and organochlorine pesticides (OCPs) are representative compounds of persistent organic pollutants (POPs). Environmental contamination by POPs is a global concern because these compounds can be accumulated and amplified through the food web and can cause adverse health effects such as development toxicity, cancer and endocrine disruption (Peterson *et al.*, 1993; Baccarelli *et al.*, 2004). Although the use of PCBs and OCPs has been banned or restricted in Korea, these contaminants are still present in coastal environment (Kim *et al.*, 2002; Hong *et al.*, 2006; Moon *et al.*, 2008). Polybrominated diphenyl ethers (PBDEs) have been widely used as brominated flame retardants (BFRs) in many products, such as TV, radio, computers and textiles (Pijnenburg *et al.*, 1995). Limited data are available concerning the concentrations and accumulations of organochlorines (OCs) and PBDEs in marine biota from Korean coastal waters (Moon and Ok, 2006; Moon *et al.*, 2007; Moon and Choi, 2009; Moon *et al.*, 2009).

Marine mammals such as cetaceans have high trophic level in food chain and relatively low metabolic activities, thus these species have increased levels of POPs and related compounds (Aguilar *et al.*, 2002; Tanabe, 2002; Kannan *et al.*, 2005; Hickie *et al.*, 2007). Bioaccumulation and long-term exposure of these contaminants may pose a threat to the health and reproductive potential of cetaceans in marine ecosystem (Jepson *et al.*, 2005; Hickie *et al.*, 2007).

National Fisheries Research and Development Institute (NFRDI) have launched monitoring of POPs in Korean coastal waters and these contaminants were determined in a wide variety of environmental matrices such as atmosphere, water, sediment and biota (Moon *et al.* 2005; 2007; 2008; Moon and Ok, 2006; Choi *et al.*, 2009). However, there has no data concerning POPs in marine mammals from Korean

coastal environment. The objective of this study was to make representative data concerning chlorinated and brominated organic compounds in cetaceans and to compare the present contamination status of these contaminants in Korean coastal waters with other countries or locations.

MATERIALS AND METHODS

Liver and blubber samples were obtained from two cetacean species, minke whale (*B. acutorostrata*) and common dolphin (*Delphinus* spp.) entangled in fishing gear such as set net along the Korean coasts in 2006. After biometric measurement for collected cetaceans, the specimens were dissected and immediately transported to the laboratory of CRI. All of the samples were kept in a freezer at -20°C until extraction.

Twenty-two PCB congeners (PCB 8, 18, 28, 29, 44, 52, 87, 101, 105, 110, 118, 128, 138, 153, 170, 180, 187, 194, 195, 200, 205 and 206) and 13 organochlorine pesticides; α -, β -, γ -HCH, hexachlorobenzene (HCB), *o*-, *p*-DDD, *p*-, *p*-DDD, *p*-, *p*-DDE, *p*-, *p*-DDT, *oxy*-chlordane, *trans*-chlordane, *cis*-chlordane, *trans*-nonachlor, and *cis*-nonachlor were analyzed in the livers and blubbers of cetaceans collected from Korean coastal waters. Twenty three PBDE congeners (BDE 17, 28, 47, 49, 66, 71, 85, 99, 100, 119, 126, 138, 153, 154, 156, 183, 184, 191, 196, 197, 206, 207 and 209) were also measured. Preparation procedures and instrumental analyses of PCBs, OCPs and PBDEs in cetaceans were performed following the methods described elsewhere (Kannan *et al.*, 2008; Moon *et al.*, 2007; Moon *et al.*, 2009), with some modifications.

RESULTS AND DISCUSSION

Concentrations of PCBs, OCPs and PBDEs in the livers and blubbers of two cetacean species from Korean coastal waters are summarized in Table 1. OCs and PBDEs were detected in all of the cetacean samples. Among the OCs analyzed, the levels of PCBs and DDTs were predominant in all of the liver and blubber samples because of their higher bioaccumulative properties and wide usage (Kim and Smith, 2001). The concentrations of CHLs, HCHs and HCB were lower than one or two orders of magnitude of concentrations of PCBs and DDTs. This contamination pattern is similar to those reported in other countries in cetaceans from Asian coastal waters (Ramu *et al.*, 2005; Kajiwara *et al.*, 2006a, b; Isobe *et al.*, 2009). The concentrations of PCBs and OCPs in cetacean samples were higher than those in fish and shellfish from Korean coastal waters (Moon and Ok, 2006; Moon and Choi, 2009; Moon *et al.*, 2009), due to the characters of biomagnification and bioaccumulation of these contaminants in marine food web. The concentrations of PBDEs in the livers and blubbers were lower than those of PCBs and DDTs.

The concentrations of PCBs, DDTs, CHLs, HCHs, HCB and PBDEs in the blubbers of minke whale and common dolphins from Korean coastal waters were compared to those reported in other locations and countries (Karlson *et al.*, 2000; Minh *et al.*, 2000; Andersen *et al.*, 2001; Borrel *et al.*, 2001; Hobbs *et al.*, 2003; Metcalfe *et al.*, 2004; Struntz *et al.*, 2004; Law *et al.*, 2005; Ramu *et al.*, 2005; Kajiwara *et al.*, 2006a, 2006b; Krahn *et al.*, 2007; Isobe *et al.*, 2009; Weijs *et al.*, 2009) (Figure 2). A variety of cetacean species from other countries or locations showed wide ranges of the concentrations of OCs and PBDEs. As shown in Figure 2, the overall contamination of organohalogen compounds in cetaceans seems to be relatively lower or similar to the concentrations of these contaminants measured from other countries. This is consistent with contamination status by the OCs and PBDEs in environmental compartments such as air, sediments and biota from Korean coastal waters (Moon *et al.* 2005; Moon and Ok, 2006; Moon *et al.*, 2007; Moon and Choi, 2009; Moon *et al.*, 2009).

We compared the concentrations of PCBs, OCPs and PBDEs in the livers and blubbers between minke whale and common dolphin (Figure 2). The concentrations of PCBs, DDTs, CHLs and PBDEs in the livers and blubbers of common dolphin were significantly ($p < 0.05$) higher than those measured in minke whale. This result can be explained by the difference of inhabitation and diet. The common dolphin is a near-shore species feeding in coastal waters, whereas minke whale migrates both through near-shore and off-shore waters. The major diets of common dolphin are long-lived and larger predatory fish with high lipid content such as herring and mackerel. The result indicates that inhabitation and diet can be important factors governing bioaccumulation and health risk to cetaceans by POPs and PBDEs in marine ecosystems. Although the overall concentrations of HCHs and HCB in the livers and blubbers of minke whale were higher than those of minke whale, the gender difference for these contaminants was not

significant.

The difference of PCBs, OCPs and PBDEs for two cetacean species according to gender was investigated (Figure 3). Although the average concentrations of PCBs, OCPs and PBDEs in male samples were higher than those of female samples, only DDTs and HCHs in the livers and HCHs in the blubbers of minke whale had significant differences between male and female. In addition, common dolphin did not show a gender difference concerning accumulation status. In general, the lactation and reproduction have been known as the major elimination process from female bodies (Metcalf *et al.*, 2004; Weijs *et al.*, 2009). Due to limited sample size of the present study and confounding inhabiting character of both species in Korean coastal waters, it was difficult to conclude whether the gender differences of PCBs, OCPs and PBDEs followed those of other studies. Further investigation of these questions is needed.

Chemical profiles of PCBs, OCPs and PBDEs in the livers and blubbers of both species from Korean coastal waters are shown in Figure 4. The profiles of PCBs and PBDEs in the livers and blubbers were similar to each other. The predominant congener of PCBs was PCB 153, followed by PCB 138, PCB 118, PCB 180 and PCB 101, which are consistent with reports for fish (Moon *et al.*, 2009) and human serum (Kang *et al.*, 2008) in Korea. The predominant PBDE congener was BDE 47, followed by BDE 154, BDE 99 and BDE 100. Deca-BDE (BDE 209) was detected in some liver and blubber samples of cetaceans from Korean coastal waters, consistent with a high consumption of deca-BDE for the flame-retardant market in Korea (Moon *et al.*, 2007). Although the accumulation patterns of OCPs in both cetaceans were different, the predominant OCP was *p,p'*-DDE in all of the liver and blubber samples.

Some studies have reported that the reproductive impairments and declining populations in marine mammals have associated with high burdens of organic pollutants (DeLong *et al.*, 1973; Helle *et al.*, 1976; Martineau *et al.*, 1987; Subramanian *et al.*, 1987; Kannan *et al.*, 1993; Corsolini *et al.*, 1995; De Swart *et al.*, 1996). The concentrations of PCBs and DDTs in the blubbers of cetaceans from Korean coastal waters were compared with the concentrations of PCBs and DDTs in other marine mammals, which showed adverse effects (Figure 5). As shown in Figure 5, the concentrations of PCBs and DDTs in the present study can have adverse effects such as low sex hormone and suppression of the immune system. Continuous monitoring and ecotoxicological studies are needed to understand the status of organic contamination as well as actual health effects of anthropogenic chemicals on this population of cetaceans.

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Table 1. Concentrations (ng/g lipid weight) of chlorinated and brominated organic pollutants in the livers and blubbers of cetaceans collected from Korean coastal waters

	Liver samples						Blubber samples					
	<u>Minke</u>			<u>Common</u>			<u>Minke</u>			<u>Common</u>		
	<u>whale</u>		Range	<u>dolphin</u>		Range	<u>whale</u>		Range	<u>dolphin</u>		Range
	Mean \pm SD	Median		Mean \pm SD	Median		Mean \pm SD	Median		Mean \pm SD	Median	
PCBs	760 \pm 980	340	140–4100	3700 \pm 2000	3950	240–8400	1980 \pm 1600	1380	390–6160	15000 \pm 7500	16000	1100–27000
DDTs	320 \pm 340	210	30–1320	1230 \pm 620	1280	80–2650	2600 \pm 2550	1370	270–1440	13500 \pm 6500	15000	730–23000
CHLs	40 \pm 40	20	6.4–170	90 \pm 50	90	6.1–230	300 \pm 210	200	30–800	1100 \pm 530	1220	60–1940
HCHs	170 \pm 90	140	70–430	150 \pm 50	160	60–230	530 \pm 500	330	40–2100	340 \pm 130	360	100–580
HCB	40 \pm 30	40	10–130	40 \pm 20	40	7.2–70	120 \pm 90	100	30–480	110 \pm 50	110	20–230
PBDEs	20 \pm 20	20	3.3–90	180 \pm 90	180	20–320	160 \pm 130	100	30–430	1650 \pm 720	1800	140–3070

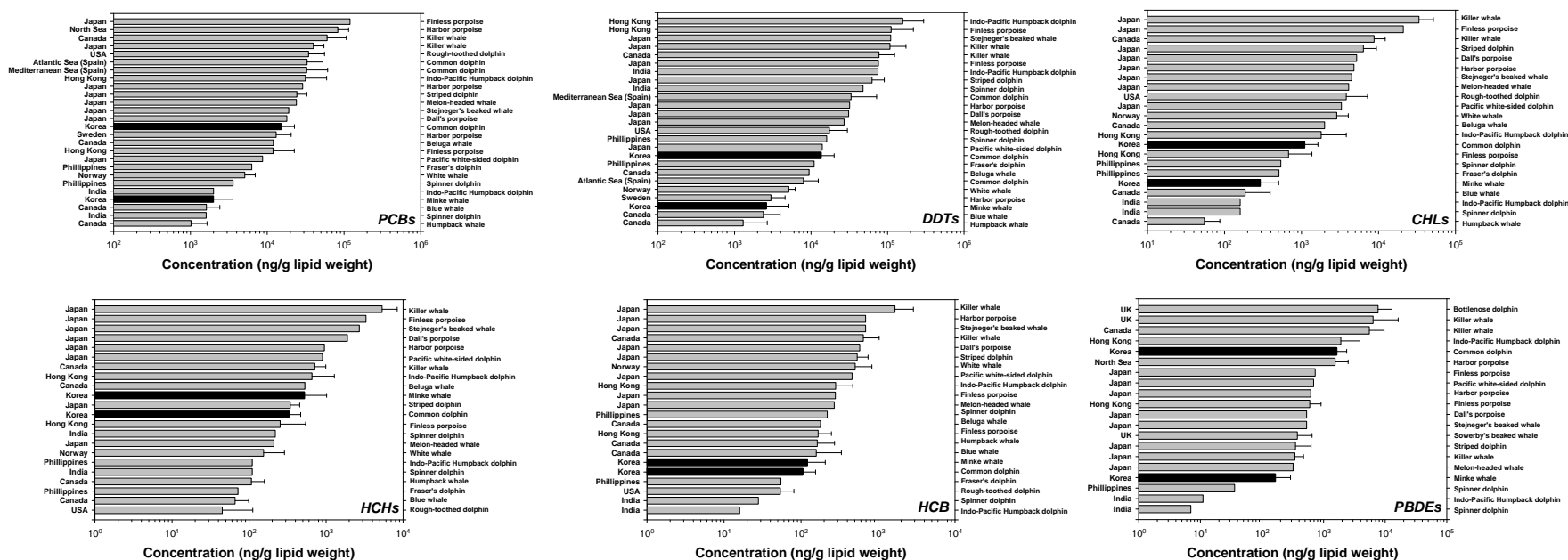


Figure 1. Comparison of average concentrations (ng/g lipid weight) of PCBs, OCPs and PBDEs in the blubbers of minke whale and common dolphin from Korean coastal waters with those reported in cetaceans from several other locations. Whiskers on the bars represent the standard deviation of individual contaminant concentration. The black boxes indicate the concentrations of PCBs, OCPs and PBDEs measured in the blubbers of cetaceans from Korean coastal waters. Compared data were obtained from Karlson et al., 2000; Minh et al., 2000; Andersen et al., 2001; Borrel et al., 2001; Hobbs et al., 2003; Metcalfe et al., 2004; Struntz et al., 2004; Law et al., 2005; Ramu et al., 2005; Kajiwarra et al., 2006a, 2006b; Krahn et al., 2007;; Isobe et al., 2009; Weijs et al., 2009.

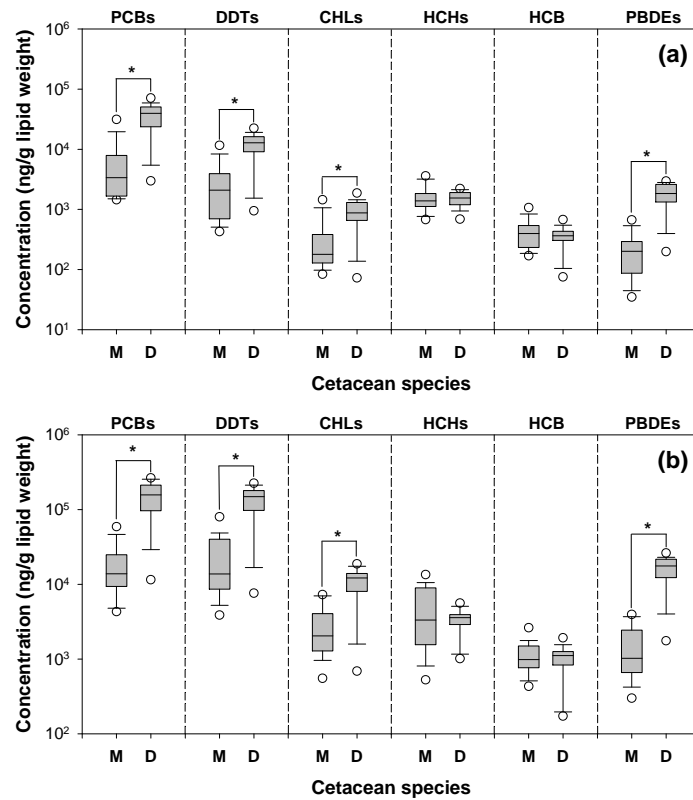


Figure 2. Comparison of the concentrations of PCBs, OCPs and PBDEs in the livers (a) and blubbers (b) of minke whales and common dolphins collected from Korean coastal waters. M represents minke whale and D represents common dolphin. The symbol of * indicates the significance at the level of $p < 0.05$ by Student t -test between two cetacean species.

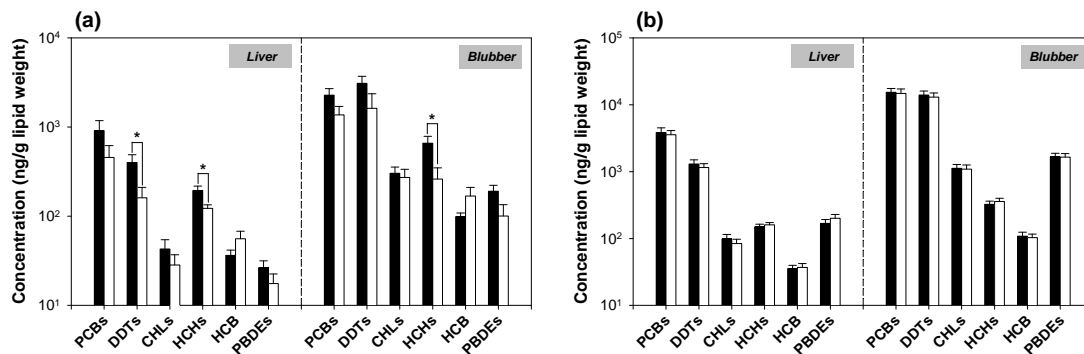


Figure 3. Comparison of the concentrations of PCBs, OCPs and PBDEs in the livers and blubbers of male and female of minke whale (a) and common dolphin (b) collected from Korean coastal waters. Whiskers on the bars represent the standard error for individual contaminants. The symbol of * indicates the significance at the level of $p < 0.05$ by Student t -test between males and females of both cetaceans.

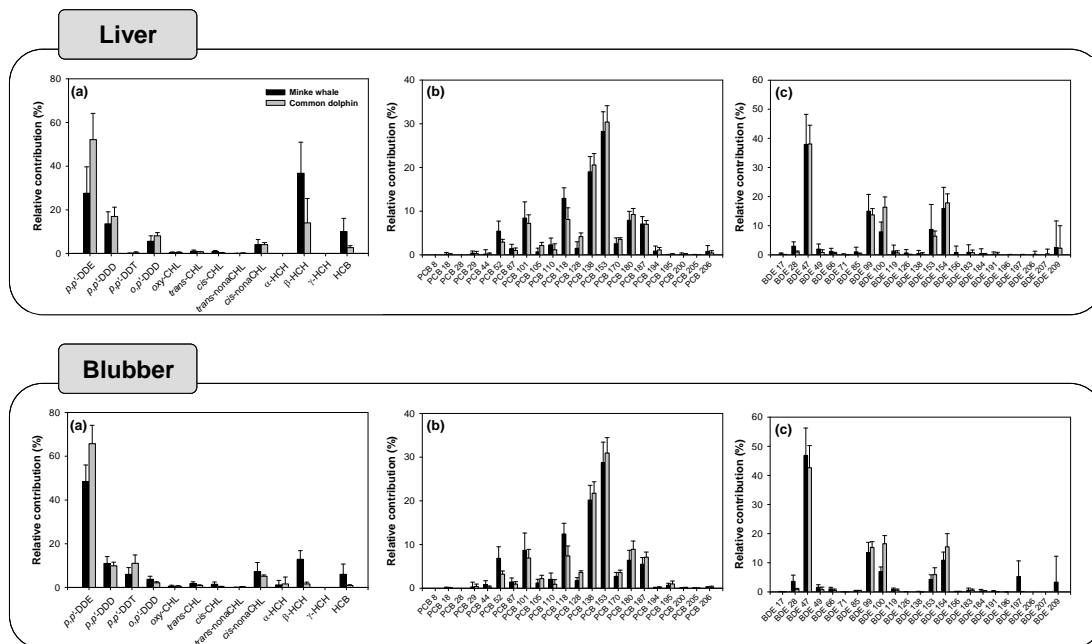


Figure 4. Relative contribution of OCPs (a), PCBs (b) and PBDEs (c) to the total concentrations of individual chemical group in the livers and blubbers of minke whale and common dolphin collected from Korean coastal waters. Whiskers on the bars represent the standard deviation for individual contribution of individual contaminants.

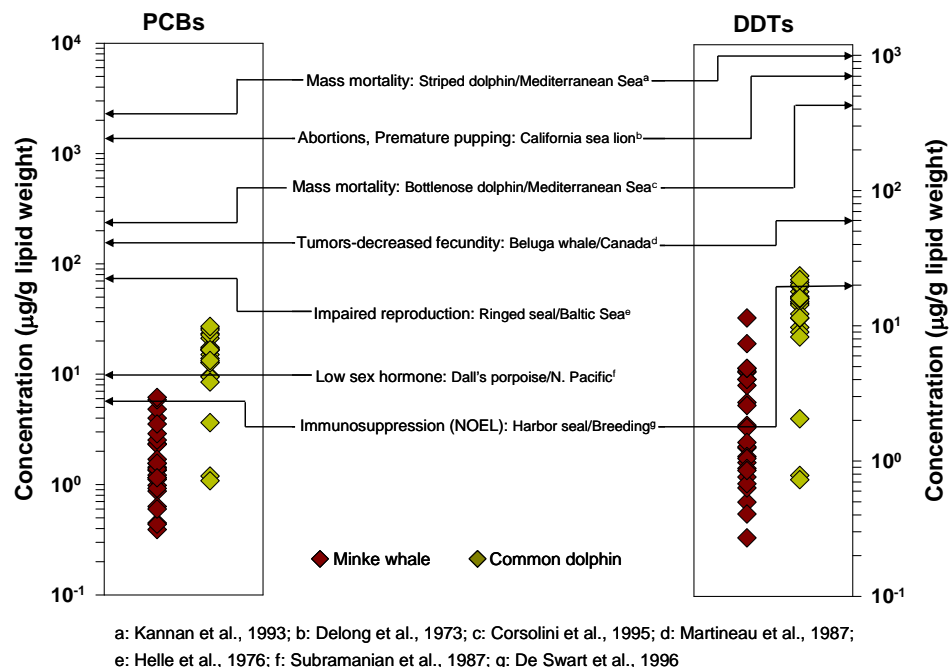


Figure 5. Toxic assessment of PCBs and DDTs in cetaceans collected from Korean coastal waters.