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## Ecotoxicology and Environmental Safety



journal homepage: www.elsevier.com/locate/ecoenv

### Persistent organochlorines in 13 shark species from offshore and coastal waters of Korea: Species-specific accumulation and contributing factors

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#### ARTICLE INFO

Article history: Received 7 January 2015 Received in revised form 9 February 2015 Accepted 10 February 2015 Available online 18 February 2015

Keywords: Organochlorines PCBs OCPs Inter-species difference Cancer risk

#### ABSTRACT

Data on persistent organochlorines (OCs) in sharks are scarce. Concentrations of OCs such as polychlorinated biphenyls (PCBs) and organochlorine pesticides (OCPs) were determined in the muscle tissue of 13 shark species (n=105) collected from offshore (Indian and Pacific Oceans) and coastal waters of Korea, to investigate species-specific accumulation of OCs and to assess the potential health risks associated with consumption of shark meat. Overall OC concentrations were highly variable not only among species but also within the same species of shark. The concentrations of PCBs, DDTs, chlordanes, hexachlorobenzene, and heptachlor in all shark species ranged from < LOO (limit of quantification) to 184 (mean: 35.0), < LOQ to 1135 (58.2), < LOQ to 56.2 (4.31), < LOQ to 18.8 (1.64) and < LOQ to 77.5 (1.37) ng/g lipid weight, respectively. The determined concentrations of PCBs and DDTs in shark in our study were relatively lower than those reported in other studies. Aggressive shark species and species inhabiting the Indian Ocean had the highest levels of OCs. Inter-species differences in the concentrations and accumulation profiles of OCs among shark species could be explained by differences in feeding habit and sampling locations. Several confounding factors such as growth velocity, trophic position, and regional contamination status may affect the bioaccumulation of OCs in sharks. Hazard ratios of non-cancer risk for all the OCs were below one, whereas the hazard ratios of lifetime cancer risks of PCBs and DDTs exceeded one, implying potential carcinogenic effects in the general population in Korea. This is the first report to document the occurrence of OCs in sharks from Korea.

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#### 1. Introduction

Organochlorines (OCs) such as polychlorinated biphenyls (PCBs) and organochlorine pesticides (OCPs) are ubiquitous contaminants on a global scale. Because of their persistent and bioaccumulative characters, OCs occur and accumulate in wildlife (Gelsleichter et al., 2005; Nakata et al., 2005; Moon et al., 2010) and humans (Bergonzi et al., 2009; Lee et al., 2013). PCBs and OCPs have adverse health effects such as developmental toxicity, cancer, and endocrine disruption (Beard, 2006; Foster et al., 2012). PCBs and OCPs are regulated by the Stockholm Convention as persistent organic pollutants (POPs) by the United Nations Environment

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http://dx.doi.org/10.1016/j.ecoenv.2015.02.021 0147-6513/© 2015 Elsevier Inc. All rights reserved. Programme (UNEP) since 2001 (Fielder et al., 2013). In Korea, approximately 4300 tons of PCBs and 3600 tons of OCPs were produced up till the 1990s (Breivik et al., 2002). Although the environmental levels of PCBs and OCPs have decreased considerably over the past three decades (Isobe et al., 2009; Sericano et al., 2014), these contaminants are still present in wildlife and human (Corsolini et al., 1995; Storelli and Marcotrigiano, 2001; Davis et al., 2002; Park et al., 2010; Lee et al., 2013).

Sharks are cartilaginous fish and usually consume plankton, mollusks, crustaceans, and bony fishes (Parker, 2008). Some aggressive sharks (e.g. blue and white sharks) hunt large fishes, seabirds, marine mammals, and even other sharks (Parker, 2008). Due to their high trophic position, long life-span, and relatively low metabolic capacity, sharks contain high levels of POPs (Johnson-Restrepo et al., 2005). However, few studies are available on accumulation of OCs in sharks, with only one or few species and small sample sizes (Corsolini et al., 1995; Storelli and Marcotrigiano, 2001; Strid et al., 2007; Cascaes et al., 2014; Lu et al., 2014; Olin et al., 2014). In some countries, such as Australia, China, Japan, and Korea, sharks are consumed by the general population in form of fillets, fin soup, and nutritional supplements (e.g. liver oil). Previous studies have confirmed the human health risks associated with consumption of shark meat (Holtcamp, 2012; Man et al., 2014). In fact, the United States Environmental Protection Agency (US EPA) designated sharks as high mercury-containing fishes, which are hazardous to infants and pregnant women (US EPA, 1997). Although OCs ingested by humans via shark consumption are likely to pose health risks, little is known about residue levels and accumulation patterns of OCs in sharks (Zhou et al., 2013).

In the present study, we determined the concentrations and accumulation features of OCs in 13 shark species collected from offshore and coastal waters of Korea, to investigate species-specific accumulation of these contaminants. The relationships between OC levels and certain biological factors (e.g. body length, body weight, lipid content, and trophic position) were investigated, to elucidate the contributing factors governing the bioaccumulation of OCs in a variety of sharks. Moreover, potential health risks to humans were assessed based on determining the daily intake of OCs due to consumption of shark meat by the general population of Korea.

#### 2. Materials and methods

#### 2.1. Sample collection

Dorsal muscles were collected from 13 shark species (n=105) found entangled in the long lines of commercial fisheries or bycatch from offshore trawls of the Indian and Pacific Oceans and coastal waters of Korea during July to October in 2010. All shark species surveyed in this study are globally endangered; they are on the red list of the International Union for Conservation and Natural Resources (IUCN) (Parker, 2008). Biological information such as body length, body weight, feeding habits, and lipid content of each species are summarized in Table 1. However, the detailed information on the sampling locations of sharks collected from seawater zones (Indian and Pacific Oceans and coastal waters) are not available in our study. After dissection of the sharks on commercial ships, the collected dorsal tissues were transported to the National Fishery Products Quality Management Service, Korea, and kept frozen at -20 °C until analysis.

#### 2.2. Experimental procedures

Twenty two PCB congeners (CBs 8, 18, 28, 29, 44, 52, 87, 101, 105, 110, 118, 128, 138, 153, 170, 180, 183, 194, 195, 200, 205 and 206) composed of tri- to nona-CBs and 16 OCP compounds were measured in shark tissues. For OCPs, six dichlorodiphenyltrichloroethanes (DDTs; *o,p'*-DDE, *p,p'*-DDE, *o,p'*-DDD, *p,p'*-DDD, *o,p* 

#### Table 1

Detailed information on the 13 shar	species collected from offshore	(Indian and Pacific Oceans	) and coastal waters of Korea.

Species (Scientific name)	nª	Sampling location	Body length (cm)	Body weight (kg)	Lipid (%)	Habitat <sup>b</sup>	Feeding habit (Major prey) <sup>b,c</sup>
Blacktip reef shark (Carcharhinus melanopterus)	26	Pacific Ocean	$90\pm16$	$21\pm11$	$4.2\pm1.3$	Coastal, continental shelf	Aggressive carnivore (cephalopods, crustaceans, bony fishes, and marine
			(63–115)	(7.0-42)	(1.6–6.2)	pelagic (20–75 m)	birds)
Spiny dogfish (Squalus acanthias)	17	Pacific Ocean	$81\pm8.2$	$2.7\pm0.9$	$18 \pm 3.6$	Coastal, continental slope	Carnivore (squid, shrimp, crustaceans, fishes, and polychaetes)
			(62–93)	(1.5–4.5)	(12–23)	lower bathyal (50– 150 m)	
Blue shark (Prionace glauca)	15	Pacific Ocean	$112\pm20$	$22\pm8.2$	$3.5\pm1.5$	Bathyal (150–350 m)	Aggressive carnivore (sardine, squid,
			(70–144)	(12-36)	(1.4–5.2)		marine birds, and marine mammals)
Pelagic thresher shark (Alopias pelagicus)	13	Pacific Ocean	$96\pm27$	$40\pm29$	$4.4\pm2.1$	Upper bathyal ( > 150 m)	Carnivore (squid and pelagic small fishes)
			(53-143)	(7.5–108)	(1.4–9.0)		
Shortfin mako (Isurus oxyrinchus)	7	Pacific Ocean	$118\pm9.8$	$44\pm26$	$4.3\pm1.4$	Coastal, bathyal ( > 500 m)	Aggressive carnivore (bony fishes, mar- ine birds, marine mammal, and sharks)
		Indian Ocean	(106–133)	(19-96)	(2.1 - 5.9)		
Cloudy dogfish (Scyliorhinu torazame)	5	Korean coast	$33\pm2.9$	$\textbf{0.8} \pm \textbf{1.0}$	$\textbf{4.8} \pm \textbf{3.4}$	Coastal, continental shelf, bathyal	Carnivore (squid, crustaceans, and small fishes)
			(30-35)	(0.2 - 2.0)	(1.7–9.7)	-	
Oceanic whitetip shark (Carch-	4	Pacific Ocean	$85 \pm 25$	$17\pm16$	$4.1\pm1.4$	Coastal, continental	Aggressive carnivore (pelagic cephalo-
arhinus longimanus)			(60–119)	(5.1–40)	(2.2–5.4)	shelf ( > 150 m)	pods, bony fishes, marine birds, and marine mammals)
Shortnose spurdog (Squalus	4	Korean coast	$87\pm2.9$	$4.2\pm0.7$	$3.4\pm2.1$	Continental shelf,	Carnivore (squid, shrimp, crustaceans,
megalops)			(85–91)	(3.5-5.1)	(1.7-6.2)	bathyal ( > 50–750 m)	and small fishes)
Milk shark (Rhizoprionodon	4	Korean coast	$108 \pm 9.6$	$9.6\pm0.7$	$3.7\pm2.1$	Continental shelf,	Carnivore (squid, crustaceans, and
acutus)			(100–120)	(9.0–11)	(1.7–6.1)	bathyal (200 m)	fishes)
Smooth hammerhead (Sphyrna zygaena)	3	Pacific Ocean Korean coast	$105\pm17$	$17 \pm 12$	$4.5\pm0.3$	Continental shelf ( > 280 m)	Aggressive carnivore (squid, crusta- ceans, bony fishes, rays, and sharks)
			(87–120)	(6.2–30)	(4.2-4.8)		
Banded houndshark (Triakis scyllium)	3	Pacific Ocean	$111\pm40$	$5.7\pm3.2$	$4.6\pm1.0$	Coastal, sandy, muddy water	Carnivore (squid, crustaceans, bony fishes, and benthic invertebrates)
			(70-150)	(2.0 - 8.0)	(3.6 - 5.6)		
Crocodile shark (Pseudocarcharias	2	Indian Ocean	$76 \pm 0.7$	$1.6 \pm 0.4$	$4.4 \pm 0.8$	Bathyal ( > 300 m)	Carnivore (squid, shrimp, and pelagic
kamoharai)			(75-76)	(1.3–1.8)	(3.9 - 5.0)		bony fishes)
Starspotted smooth-hound (Mus- telus manazo)	2	Korean coast	60	$1.6\pm0.8$	6.0 ± 1.0	Coastal, sandy, muddy waters (200–360 m)	Carnivore (squid, shrimp, crustacean, shellfish, and small fishes)
-				(1.0-2.2)	(5.3–6.8)		-

<sup>a</sup> Sample number analyzed.

<sup>b</sup> Parker, 2008.

<sup>c</sup> Stillwell and Kohler, 1982.

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