Decreases in Concentrations and Human Dietary Intakes of Polychlorinated Biphenyls (PCBs) and Polybrominated Diphenyl Ethers (PBDEs) in Korean Seafood Between 2005 and 2017

Minkyu Choi
National Institute of Fisheries Science
In-seok Lee (islee@korea.kr)
National Institute of Fisheries Science https://orcid.org/0000-0002-6334-8405

Original Research

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Abstract

Concentrations of PCBs and PBDEs were measured in 23 seafood species widely consumed by Korean population in 2005–2007, 2010–2011, and 2015–2017. The Σ_{82}PCB and Σ_{19}PBDE concentrations in the seafood samples of 2015–2017 were 0.06–6.69 ng/g wet weight and 0.01–1.60 ng/g wet weight, respectively. The Σ_{82}PCB and Σ_{19}PBDE concentrations in the samples were correlated significantly, and elevated PCB and PBDE concentrations were found in fatty fish such as herring, mackerel, and tuna. The current intakes of PCBs and PBDEs were much lower than a TDI or a LOAEL. The levels and human dietary intakes of PCBs and PBDEs in the 2015–2017 survey showed decreases of 17–73% and 57–86%, respectively, in relation to the 2005–2007 and 2010–2011 surveys. This indicates that global bans on PCBs and PBDEs have been effective, and their levels and exposure have been gradually declining.

1. Introduction

Polychlorinated biphenyls (PCBs) and polybrominated diphenyl ethers (PBDEs) are persistent organic pollutants (POPs) that accumulate in organisms and are biomagnified through food chains with elevated levels in top predators (Moon et al. 2010). These contaminants can cause adverse health effects such as dermal toxicity, immunotoxicity, reproductive deficits, teratogenicity and endocrine interferences (Ankarberg et al. 2007). Use of POPs has been banned globally according to Stockholm convention on POPs, and PCBs and PBDEs were classified as POPs in 2005 and 2009, respectively (Convention 2008). In Korea, approximately 4,300 tons of PCBs were used until the 1990s, and the use of PCBs was banned (Moon et al. 2009). In Korea, PBDEs have been widely used as brominated flame retardants. Usage of deca-BDE in Korea in 2002 was 12,324 tons, while usage of penta- and octa-BDEs together was 84 tons. Penta- and octa-BDE were banned in 2008, and deca-BDE was not restricted, but according to voluntary phase out, usage of deca-BDE in Korea was decreased to 1,405 ton in 2010 (MOE 2015). As PBDEs are not bonded into materials but simply blended with the polymers, they are likely to be released into the environment and entering biological tissues through food chains. These contaminants are still found in seafood as well as coastal environments, and are considered to pose a substantial risk for human health (Leng et al. 2014). The recent concern now is to check the effectiveness of the global bans and efforts in terms of how fast the POPs such as PCBs and PBDEs in environmental levels and risks decline in the world.

A temporal trend study is an essential tool for assessing the effectiveness of legislative action on target contaminants in the environment (Jeong et al. 2016; Toms et al. 2018). However, little is known about temporal trends of dietary exposures of POPs such as PCBs and PBDEs associated with seafood consumption. In 2005–2007 and 2010–2011, we investigated, for nationwide baseline study, seafood contaminations focused on determining the concentration of POPs including PCBs and PBDEs in seafood most consumed by Korean population. Subsequently, the exposures of PCBs and dioxin-like PCBs (DL-PCBs) via seafood consumption were estimated to 4.4 ng/kg bw/day and 0.9 pg-TEQ/kg bw/day, respectively (Moon and Choi 2009; Moon et al. 2009). In 2010–2011, the exposures of PBDEs via seafood intake was estimated to 0.4 ng/kg bw/day (MLTM 2012). In order to investigate temporal trend of levels and the total dietary exposures to PCBs and PBDEs, the same seafood groups (fish, crustaceans, cephalopods, and bivalves) were again collected and analyzed from 2015 to 2017. We investigated the current levels of 82 PCBs and 19 PBDEs in seafood and their dietary intakes via seafood, and the comparison of the present situation with the previous baseline studies.

2. Materials And Methods

2.1. Sample collection

31 marine species (fish, shellfish, crustaceans, cephalopods, bivalves and gastropods) were selected among those most commonly consumed and commercially important species in Korea, and were caught in the Yellow Sea, South Sea, and East Sea areas of Korea, as in our previous nationwide survey (MLTM 2012; Moon and Choi 2009; Moon et al. 2009). 31 marine species (n = 230) were annually collected twice or three times from the Busan cooperative fish market, which is the largest fish market in South Korea and the representative market covering the whole country, along with information on fishing areas and vessels. Collected samples were stored in a cooler box with ice and immediately transported to the laboratory. After removing the skin of the fish and cephalopods, the muscles and tissues were homogenized using an ultra-disperser. The shells of the bivalves, gastropods and crustaceans were removed, and the whole soft tissues were pooled and homogenized for analysis. For PCBs and PBDEs analyses, two or three composite samples were prepared for each seafood item for each year (n = 230). Each composite sample consisted of more than 20 individual units for fish, crustaceans, cephalopods, and 100 individual units for bivalves. Only edible parts of each seafood were included in the composites.

2.2. Chemical analysis

82 PCB congeners including 12 DL-PCB congeners (CB-77, 81, 105, 114, 118, 123, 126, 156, 157, 167, 169 and 189) and 6 Non Dioxin-Like (NDL)-PCB congeners (CB-28, 52, 101, 138, 153, 180), and 19 PBDE congeners (BDE-17, 28, 47, 49, 66, 71, 77, 85, 99, 100, 119, 126, 138, 153, 154, 156, 183, 184, and 191) were analyzed in seafood samples. 82 PCB stock standard solution, 35 $^{13}C_{12}$-labelled PCBs for surrogate standards, and seven $^{13}C_{12}$-labelled PCBs for internal standards were purchased from Wellington (Guelph, ON, Canada). 19 PBDE stock standard solution, eleven $^{13}C_{12}$-labelled PBDEs for surrogate standards, and the $^{13}C_{12}$-labelled PBDE (BDE-138) for an internal standard were purchased from Wellington (Guelph, ON, Canada).

Analyses of PCBs and PBDEs in seafood were performed according to the methods described elsewhere (Moon and Choi 2009; Moon et al. 2009). The edible tissues (approximately 50 g) were digested in 200 mL of 1 N ethanolic KOH solution (Wako Pure Chemicals, Tokyo, Japan) for 2 h by mechanic shaking. The surrogate standards for PCBs and PBDEs were spiked into the samples before the digestion process. The alkaline solutions were extracted twice with 150 mL of hexane (Ultra residue analysis, J.T.Baker, Phillipsburg, NJ, USA). The extracts were washed with purified water and then dried over 50 g of anhydrous Na_2SO_4 and subsequently reduced to 10 mL by rotary evaporation. Purification of PCBs and PBDEs was performed using multi-layer silica gel column containing Na_2SO_4 anhydrous (4 g), 10% $AgNO_3$ silica gel (3 g), silica gel (0.6 g), 22% (w/w) H_2SO_4 silica gel (3 g), 44% (w/w) H_2SO_4 silica gel (4 g), silica gel (0.6 g) and 2% (w/w) KOH silica gel (2 g) with 150 mL of DCM in hexane. The eluents were concentrated to approximately 200 μL for instrumental analysis.

2.3. Instrumental analysis and quality control

Analyses of PCBs and PBDEs were performed with a gas chromatograph (GC, Agilent Technologies 6890; USA) coupled with a high resolution mass spectrometer (HRMS, JEOL 700D, Tokyo, Japan). For PCBs, the capillary column used was a DB5-MS column (60-m length, 0.25-mm inner diameter, 0.25-μm film thickness; J & W Scientific, Palo Alto, CA, USA). Helium was used as the carrier gas at a flow rate of 1.2 mL min^{-1}. The oven temperature program was 100°C for 4 min, 100 to 180°C at 20°C min^{-1}, 180°C for 10 min, 180 to 300°C at 2°C min^{-1}, 300°C for 6.3 min. The injector temperature was 280°C. One microliter of sample was injected in a splitless injection.

For PBDEs, the capillary column used was a DB5-MS column (30-m length, 0.25-mm inner diameter, 0.25-μm film thickness; J & W Scientific, Palo Alto, CA, USA). Helium was used as the carrier gas at a flow rate of 1.0 mL min^{-1}. The oven temperature program was 100°C for 1 min, 100 to 220°C at 20°C min^{-1}, 220°C for 4 min, 220 to 300°C at 8°C min^{-1}, 300°C for 3 min. The injector temperature was 260°C. The HRMS was operated in electron ionization (EI) mode, and ions of individual PCBs and PBDEs were monitored by the selective ion monitoring (SIM) mode.

The accuracy of the method was determined by analyzing Certified Reference Materials (CRM) of a fish tissue (SRM 1946), from the US National Institute for Standards and Technology (NIST; Gaithersburg, MD, USA). The mean anlalyte concentrations found in the seven CRM samples were 71–105% and 68–98% of the certified PCB and PBDE concentrations, respectively. The mean recoveries of spiked surrogate internal standards of PCBs and PBDEs were 81 ± 15%, and 84 ± 12% respectively. The calculated limits of detection (LOD, the concentration giving a signal-to-noise ratio of 3) were 0.001–0.023 ng/g for individual PCB congeners and 0.002–0.033 ng/g for individual PBDE congeners.

2.4. Calculation of dietary intakes

The intake of a grouping of PCBs or PBDEs (ng/kg body weight/day) in a seafood species by an ‘average' Korean was calculated by multiplying the mean concentration of PCBs or PBDEs grouping (ng/g wet) in seafood species of interest by the mean daily per capita intake of the seafood species in Korea (g/day) and dividing the result by mean body weight for Koreans (kg body weight). The concentrations of undetected compounds were considered as one-half of the respective LOD. Daily consumption rates of each seafood for the general population of Korea were obtained from the National Health and Nutrition Survey (MOHW 2011, 2015).

3. Results And Discussion

3.1. Concentrations and profiles of PCBs

Table 1 shows the concentrations of Σ_{82}PCB, Σ_6NDL-PCB, Σ_{12}DL-PCB, Σ_{19}PBDE, and Σ_7PBDE in 31 seafood species widely consumed in Korea, 2015–2017. The concentrations of Σ_{82}PCB were 0.06–6.69 ng/g wet, and CB-153 was present in the highest proportion (14 ± 2.9%) among PCB congeners, followed by CB-138 (7.2 ± 1.8%), CB-118 (5.6 ± 1.6%), and CB-101 (5.0 ± 1.7%). Herring, eel, tuna and Spanish mackerel presented relatively high concentrations (6.69, 4.41, 3.73, and 3.68 ng/g wet, respectively) while the species with low levels were shrimp (0.06 ng/g wet) and abalone (0.07 ng/g wet).

Table 1
Summary in concentrations of PCBs and PBDEs in seafood from Korea, 2015–2017

No	Species	Common name	Scientific name	Consumption (g/day)[a]	Lipid (%)	Σ_{82}PCB (ng/g wet)	Σ_{6}NDL-PCB[c] (ng/g wet)	Σ_{12}DL-PCB[d] (ng/g wet)	Σ_{12}DL-PCB (pg-TEQ/g wet)	Σ_{19}PBDE[e] (ng/g wet)	Σ_{7}PBDE[f] (ng/g wet)
1	Fish	Alaska pollack	*Theragra chalcogramma*	2.8 (7.6)[b]	0.6 ± 0.1	0.85 ± 0.61	0.30 ± 0.22	0.07 ± 0.08	0.065 ± 0.094	0.06 ± 0.09	0.04 ± 0.06
2	Fish	Anchovy	*Engraulis japonicus*	1.7 (4.6)	0.6 ± 0.7	1.92 ± 1.16	0.70 ± 0.42	0.27 ± 0.17	0.312 ± 0.187	0.33 ± 0.20	0.21 ± 0.14
3	Fish	Anger fish	*Lophiomus setigerus*	0.9 (2.4)	0.4 ± 0.9	0.35 ± 0.07	0.13 ± 0.03	0.04 ± 0.01	0.024 ± 0.018	0.06 ± 0.01	0.04 ± 0.01
4	Fish	Cod	*Gadus macrocephalus*	0.6 (1.6)	0.3 ± 0.3	1.02 ± 0.58	0.39 ± 0.22	0.15 ± 0.09	0.140 ± 0.073	0.20 ± 0.09	0.11 ± 0.09
5	Fish	Eel	*Conger myriaster*	2.2 (6.0)	7.5 ± 2.3	4.41 ± 1.48	1.70 ± 0.58	0.63 ± 0.23	0.679 ± 0.210	0.63 ± 0.18	0.38 ± 0.15
6	Fish	Filefish	*Stephanolepis cirrhifer*	1.1 (3.0)	2.6 ± 16	0.13 ± 0.11	0.04 ± 0.04	0.01 ± 0.01	0.002 ± 0.005	0.03 ± 0.02	0.02 ± 0.01
7	Fish	Flounder	*Pleuronectes herzensteini*	0.6 (1.6)	0.9 ± 0.3	1.92 ± 0.66	0.71 ± 0.25	0.25 ± 0.09	0.274 ± 0.082	0.41 ± 0.11	0.26 ± 0.10
8	Fish	Gizzard shad	*Konosirus punctatus*	0.1 (0.3)	5.2 ± 0.1	2.74 ± 1.88	0.95 ± 0.66	0.21 ± 0.14	0.239 ± 0.186	0.41 ± 0.23	0.18 ± 0.13
9	Fish	Hairtail	*Trichiurus lepturus*	1.0 (2.7)	5.0 ± 7.2	1.79 ± 0.64	0.61 ± 0.22	0.20 ± 0.09	0.234 ± 0.085	0.42 ± 0.15	0.28 ± 0.09
10	Fish	Herring	*Clupea pallasii*	0.1 (0.3)	3.0 ± 8.5	6.69 ± 1.55	2.23 ± 0.55	0.71 ± 0.19	0.614 ± 0.125	1.60 ± 0.60	1.04 ± 0.38
11	Fish	Mackerel	*Scomber japonicus*	4.7 (13)	3.1 ± 10	1.69 ± 0.63	0.52 ± 0.21	0.17 ± 0.07	0.381 ± 0.151	0.62 ± 0.20	0.33 ± 0.18
12	Fish	Olive flounder	*Paralichthys olivaceus*	1.1 (3.0)	0.3 ± 0.9	0.76 ± 0.47	0.26 ± 0.17	0.07 ± 0.05	0.083 ± 0.066	0.18 ± 0.12	0.11 ± 0.09
13	Fish	Puffer fish	*Takifugu rubripes*	0.5 (1.4)	0.1 ± 0.1	0.21 ± 0.11	0.07 ± 0.04	0.02 ± 0.01	0.026 ± 0.030	0.04 ± 0.01	0.02 ± 0.01
14	Fish	Ray	*Raja kenojei*	0.1 (0.3)	0.4 ± 0.3	0.22 ± 0.15	0.09 ± 0.06	0.03 ± 0.02	0.049 ± 0.049	0.02 ± 0.02	0.02 ± 0.01
15	Fish	Rockfish	*Sebastes schlegelii*	0.8 (2.2)	0.7 ± 3.4	2.10 ± 1.55	0.74 ± 0.58	0.24 ± 0.21	0.296 ± 0.183	0.49 ± 0.34	0.31 ± 0.23

a. Daily consumption rates of each seafood for the general population of Korea were obtained from the National Health and Nutrition Survey (MOHW, 2015),

b. The percentage of each species to the total seafood consumption (%),

c. Sum of 6 Non Dioxin-Like (NDL)-PCB congeners (CB-28, 52, 101, 138, 153, 180),

d. Sum of 12 DL-PCB congeners (CB-77, 81, 105, 114, 118, 123, 126, 156, 157, 167, 169 and 189),

e. Sum of 19 PBDE congeners (BDE-17, 28, 47, 49, 66, 71, 77, 85, 99, 100, 119, 126, 138, 153, 154, 156, 183, 184, and 191),

f. Sum of 7 PBDE congeners (BDE-28, 47, 99, 100, 153, 154, and 183).

No	Species	Common name	Scientific name	Consumption (g/day)[a]	Lipid (%)	Σ_{82}PCB (ng/g wet)	Σ_6NDL-PCB[c] (ng/g wet)	Σ_{12}DL-PCB[d] (ng/g wet)	Σ_{12}DL-PCB (pg-TEQ/g wet)	Σ_{19}PBDE[e] (ng/g wet)	Σ_7PBDE[f] (ng/g wet)
16	Fish	Saury	*Cololabis saira*	0.9 (2.4)	5.0 ± 2.5	1.65 ± 0.67	0.54 ± 0.22	0.19 ± 0.09	0.418 ± 0.228	0.65 ± 0.28	0.45 ± 0.19
17	Fish	Sea bass	*Lateolabrax jaonicus*	0.1 (0.3)	2.9 ± 1.8	2.22 ± 1.22	0.77 ± 0.39	0.23 ± 0.13	0.289 ± 0.212	0.57 ± 0.33	0.35 ± 0.20
18	Fish	Spanish mackerel	*Scomberomorus niphonius*	0.6 (1.6)	8.3 ± 2.2	3.68 ± 0.92	1.20 ± 0.31	0.38 ± 0.14	0.721 ± 0.246	1.00 ± 0.26	0.50 ± 0.21
19	Fish	Tuna	*Thunnus thynnus*	0.8 (2.2)	0.4 ± 0.6	3.73 ± 2.91	1.32 ± 1.05	0.43 ± 0.36	0.475 ± 0.348	0.80 ± 0.60	0.53 ± 0.39
20	Fish	Yellow croaker	*Larimichthys polyactis*	1.7 (4.6)	3.4 ± 4.4	0.54 ± 0.19	0.16 ± 0.06	0.05 ± 0.02	0.046 ± 0.029	0.14 ± 0.06	0.08 ± 0.05
21	Crustaceans	Crab	*Portunus trituberculatus*	1.7 (4.6)	1.2 ± 5.7	0.13 ± 0.04	0.05 ± 0.02	0.01 ± 0.01	0.026 ± 0.017	0.03 ± 0.01	0.02 ± 0.01
22	Crustaceans	Shrimp	*Solenocera melantho*	2.2 (6.0)	0.6 ± 0.1	0.06 ± 0.02	0.02 ± 0.01	0.01 ± 0.01	0.001 ± 0.001	0.01 ± 0.01	0.01 ± 0.01
23	Gastropods	Abalone	*Nordotis discus*	0.4 (1.1)	1.1 ± 1.0	0.07 ± 0.10	0.03 ± 0.04	0.01 ± 0.01	0.004 ± 0.010	0.02 ± 0.03	0.01 ± 0.02
24	Bivalves	Ark shell	*Scapharca subcrenata*	0.5 (1.4)	6.1 ± 1.7	0.15 ± 0.03	0.04 ± 0.01	0.01 ± 0.01	0.086 ± 0.016	0.02 ± 1.95	0.02 ± 0.01
25	Bivalves	Clam	*Tapes philippinarum*	1.2 (3.3)	3.6 ± 0.7	0.21 ± 0.11	0.07 ± 0.04	0.01 ± 0.01	0.017 ± 0.020	0.04 ± 0.04	0.03 ± 0.03
26	Bivalves	Mussel	*Mytilus edulis*	1.3 (3.5)	7.2 ± 1.7	0.37 ± 0.20	0.14 ± 0.08	0.03 ± 0.02	0.113 ± 0.077	0.09 ± 0.06	0.07 ± 0.05
27	Bivalves	Oyster	*Crassostrea gigas*	0.9 (2.4)	5.9 ± 1.1	0.49 ± 0.18	0.16 ± 0.06	0.03 ± 0.01	0.203 ± 0.119	0.14 ± 0.13	0.09 ± 0.07
28	Cephalopods	Giant octopus	*Enteroctopus dofleini*	0.6 (1.6)	0.3 ± 0.1	0.21 ± 0.07	0.08 ± 0.03	0.03 ± 0.01	0.037 ± 0.021	0.02 ± 0.01	0.02 ± 0.01
29	Cephalopods	Octopus	*Octopus minor*	2.0 (5.4)	0.6 ± 0.5	0.18 ± 0.06	0.07 ± 0.02	0.02 ± 0.01	0.020 ± 0.024	0.03 ± 0.01	0.02 ± 0.01
30	Cephalopods	Webfoot octopus	*Octopus ocellatus*	0.2 (0.5)	0.6 ± 0.4	0.21 ± 0.21	0.07 ± 0.07	0.02 ± 0.02	0.020 ± 0.016	0.04 ± 0.04	0.03 ± 0.03

a. Daily consumption rates of each seafood for the general population of Korea were obtained from the National Health and Nutrition Survey (MOHW, 2015),

b. The percentage of each species to the total seafood consumption (%),

c. Sum of 6 Non Dioxin-Like (NDL)-PCB congeners (CB-28, 52, 101, 138, 153, 180),

d. Sum of 12 DL-PCB congeners (CB-77, 81, 105, 114, 118, 123, 126, 156, 157, 167, 169 and 189),

e. Sum of 19 PBDE congeners (BDE-17, 28, 47, 49, 66, 71, 77, 85, 99, 100, 119, 126, 138, 153, 154, 156, 183, 184, and 191),

f. Sum of 7 PBDE congeners (BDE-28, 47, 99, 100, 153, 154, and 183).

No	Species	Common name	Scientific name	Consumption (g/day)[a]	Lipid (%)	Σ_{82}PCB (ng/g wet)	Σ_6NDL-PCB[c] (ng/g wet)	Σ_{12}DL-PCB[d] (ng/g wet)	Σ_{12}DL-PCB (pg-TEQ/g wet)	Σ_{19}PBDE[e] (ng/g wet)	Σ_7PBDE[f] (ng/g wet)
31	Cephalopods	Squid	*Todaroldes pacificus*	6.2 (17)	1.3 ± 0.5	0.70 ± 0.15	0.23 ± 0.05	0.09 ± 0.02	0.082 ± 0.015	0.09 ± 0.02	0.07 ± 0.02

a. Daily consumption rates of each seafood for the general population of Korea were obtained from the National Health and Nutrition Survey (MOHW, 2015),

b. The percentage of each species to the total seafood consumption (%),

c. Sum of 6 Non Dioxin-Like (NDL)-PCB congeners (CB-28, 52, 101, 138, 153, 180),

d. Sum of 12 DL-PCB congeners (CB-77, 81, 105, 114, 118, 123, 126, 156, 157, 167, 169 and 189),

e. Sum of 19 PBDE congeners (BDE-17, 28, 47, 49, 66, 71, 77, 85, 99, 100, 119, 126, 138, 153, 154, 156, 183, 184, and 191),

f. Sum of 7 PBDE congeners (BDE-28, 47, 99, 100, 153, 154, and 183).

The levels of Σ_6NDL-PCB were 0.02–2.23 ng/g wet, and CB-153 contributed more than 41% to the Σ_6NDL-PCB (Fig. 1). The levels of Σ_6NDL-PCB contributed 35% to the Σ_{82}PCB, and the levels of Σ_6NDL-PCB were strongly correlated with those of Σ_{82}PCB ($r = 0.997$, $p < 0.01$). Herring, eel, tuna, and Spanish mackerel also showed the highest concentrations of Σ_6NDL-PCB. None of the samples showed concentrations of Σ_6NDL-PCB above the European Union threshold concentrations, set at 75 ng/g wet for fish and 300 ng/g wet for eels (Official Journal of the European Union 2011). The levels of Σ_6NDL-PCB in this study was lower than those in Polish Baltic fishing area (Piskorska-Pliszczynska et al. 2012), Catalonia of Spain (Perelló et al. 2015), four areas (South China Sea, Bohai Sea, East China Sea, and Yellow Sea) of China (Liu et al. 2011; Qian et al. 2017), three areas (Adriatic, Ionian, and Tyrrhenian seas) of Italy (Miniero et al. 2014) and nationwide survey of France (Arnich et al. 2009), shown in Table 2.

Table 2
The concentrations of PCBs and PBDEs in seafood from many countries

Country	Species	Sampling year	Σ_6 NDL-PCB[a] (ng/g wet)	Σ_{12} DL-PCB[b] (pg-TEQ/g wet)	Σ_7 PBDE[c] (ng/g wet)	Reference
Poland	5 Fish species (n = 177)	2006–2010	1.11–38.7	0.64–6.07		Piskorska-Pliszczynska et al. (2012)
Sweden	13 Fish/fish products	2005		0.177–0.355		Törnkvist et al. (2011)
Spain	14 Seafood species 14 Seafood species (n = 384)	2006 2012	2.16–49.95	0.17–1.99	0.01-1.62[d]	Domingo et al. (2008), Perelló et al. (2015)
France	Seafood (n = 1,119)	2002–2006	0.03-1,157			Arnich et al. (2009)
Italy	14 Fish/crustacean species (n = 40)	2007–2008	0.768-148	0.027–11.8	0.06–2.98	Miniero et al. (2014)
US	18 Fish species (n = 470)	2006–2013			1-390[e] (1-300)[f]	Gandhi et al. (2017)
Japan	9 Fish/shellfish (n = 18)	1998		0.033–7.24		Naito et al. (2003)
China	30 Fish species (n = 44) 17 Seafood species (n = 289)	- 2014	0.3-2,972 <LOD-64		0.2–476	Liu et al. (2011), Qian et al. (2017)
Australia	24 Seafood species (n = 24)	-			0.61-21.39[d]	Shanmuganathan et al. (2011)
East Africa	2 Fish species (n = 64)	2011		0.001–0.02		Ssebugere et al. (2013)
Korea	31 Seafood species (n = 230)	2015–2017	0.04–2.23	0.002–0.721	0.01–1.04	This study

a. Sum of 6 Non Dioxin-Like (NDL)-PCB congeners (CB-28, 52, 101, 138, 153, 180),

b. Sum of 12 DL-PCB congeners (CB-77, 81, 105, 114, 118, 123, 126, 156, 157, 167, 169 and 189),

c. Sum of 7 PBDE congeners (BDE-28, 47, 99, 100, 153, 154, and 183),

d. Sum of 6 PBDE congeners (BDE-47, 99, 100, 153, 154, and 183),

e. Sum of 33 PBDE congeners,

f. BDE-47.

The levels of Σ_{12}DL-PCB were 0.01–0.71 ng/g wet and CB-118 contributed 55% to Σ_{12}DL-PCB. The levels of Σ_{12}DL-PCB contributed 10% to the Σ_{82}PCB, and the levels of Σ_{12}DL-PCB were significantly correlated with those of Σ_{82}PCB (r = 0.980, $p < 0.01$). Based on the WHO-TEF values (2005), Σ_{12}DL-PCB concentrations were 0.001–0.721 pg-TEQ/g wet, and the dominant congeners were CB-126 (89 ± 6.2%), which has the highest toxic equivalent factor (TEF 0.1) among DL-PCBs, followed by CB-169 (8.4 ± 6.3%). Spanish mackerel (0.721 pg-TEQ/g wet), eel (0.679 pg-TEQ/g wet), herring (0.614 pg-TEQ/g wet), and tuna (0.475 pg-TEQ/g wet) showed high concentrations of DL-PCBs, similarly to NDL-PCBs. The European Union threshold concentrations of sum dioxins (PCDD/Fs + DL-PCBs) have been set at 6.5 pg-TEQ/g wet for fish and 10 pg-TEQ/g wet for eels (Official Journal of the European Union 2011). The contributions of four species (Spanish mackerel, eel, herring and tuna) to the threshold concentrations were 6.8–11%, indicating that DL-PCB levels in seafood from Korea were safe. The levels of DL-PCBs in the present study were comparable to or lower than those in four major cities (Malmoe, Gothenburg, Uppsala and Sundsvall) of Sweden (Törnkvist et al. 2011), and Catalonia of Spain (Perelló et al. 2015), Ariake Sea of Japan (Naito et al. 2003), three areas (Adriatic, Ionian, and Tyrrhenian seas) of Italy (Miniero et al. 2014), and Polish Baltic fishing area (Piskorska-Pliszczynska et al. 2012), shown in Table 2.

3.2. Concentrations and profiles of PBDEs

The concentrations of Σ_{19}PBDE were 0.01–1.60 ng/g wet, and herring (1.60 ng/g wet) was the species showing the highest PBDE level, followed by Spanish mackerel (1.00 ng/g wet) and tuna (0.80 ng/g wet) (Table 1). The Σ_{19}PBDE and Σ_{82}PCB concentrations in the samples correlated significantly (r = 0.952, $p < 0.01$). The result agrees with the findings in fish species and aquatic bird eggs (Liu et al. 2011; Manchester-Neesvig et al. 2001). Manchester-Neesvig et al. (2001) found that the concentrations of PBDEs and PCBs are highly correlated in individual fish, implying that PBDEs are as prevalent as PCBs in Lake Michigan. Liu et al. (2011) found that PBDEs have been in Chinese marine environment for many years and

PBDE congeners (except for BDE-209), like PCBs, were distributed throughout the individual marine areas. This indicates that PBDEs, along with PCBs, were distributed throughout the coastal waters and have a similar accumulation pattern in seafood species.

The PBDE patterns in seafood and marine mammals are usually dominated by penta-mix formulation related congeners (Law et al. 2006; Moon et al. 2010). In the present study, Σ_7PBDE (BDE-28, 47, 99, 100, 153, 154 and 183), which were related to PBDE Penta-mix (BDE-47, 99, 100, 153 and 154) and Octa-mix (BDE-183), showed the ranges of 0.01–1.04 ng/g wet, and contributed 66 ± 9.3% to Σ_{19}PBDE, and the levels of Σ_7PBDE were significantly correlated with those of Σ_{19}PBDE (r = 0.990, $p < 0.01$). It was found that the PBDE profile in seafood was BDE-47 (42%) > BDE-154 (23%) + BDE-153 (6.7%) > BDE-100 (16%) + BDE-99 (7.9%) > BDE-28 (4.4%) > BDE-183 (0.8%) (Fig. 1). PBDE profiles in fish were generally as follows: BDE-47 > BDE-99 + BDE-100 > BDE-153 + BDE-154 (Hites 2004). In fishes from Europe, these five congeners accounted for 69%, 28%, and 8% of ΣPBDE, respectively (Hites 2004). These profiles did not reflect the composition of penta-mix formulations, which contained 24–38% tetra-BDEs (BDE-47), 50–60% penta-BDEs (BDE-99 and 100), and 4–8% hexa-BDEs (BDE-153 and 154; de Wit 2002). This might be associated with the differences in the uptake and removal efficiency of particular congeners. In uptake studies of PBDEs to pike (Burreau et al. 2004), BDE-47 was characterized by the highest uptake efficiency (90% of the given dose), followed by BDE-99 (62%) and BDE-153 (40%). Bioaccumulation studies of PBDEs in zebrafish also showed the highest accumulation of BDE-47 (de Wit 2002). In this study, BDE-154 was also found at relatively higher proportions than BDE-99 and BDE-100 in seafood species. It is similar to those in seafood from China (Xia et al. 2011). The reasons for the relative high proportion of BDE-154 may be due to the extensive use of Octa-BDE rather than Penta-BDE and the debromination of higher brominated congeners, BDE-183 to BDE-154 in fish species (Xia et al. 2011). In Korea, usage of octa-BDE (179 ton) in 2003 was 10 times higher than usage of penta-BDEs (19 ton) (MOE 2015).

The concentrations of Σ_7PBDE (0.01–1.04 ng/g wet) in seafood in our study were similar to those reported from Catalonia of Spain (0.012–1.62 ng/g wet, Domingo et al. 2008) and Italy (0.06-2.98ng/g wet; Miniero et al. 2014), but were lower than those reported from China (0.2–476 ng/g wet, Liu et al., 2011), US (1-300 ng/g wet; Gandhi et al. 2017), and Australia (0.61–21.39 ng/g wet; Shanmuganathan et al. 2011).

3.3. Human exposure from seafood consumption

Table 3 shows the exposure of total PCBs and PBDEs by Korean population consuming seafood. The body weight, 59.8 kg, was used to calculate the daily intakes for an average Korean. For Korean population, the dietary intakes of total PCB and 6 NDL-PCB were 45.2 ng/day (0.76 ng/kg bw/day) and 15.7 ng/day (0.26 ng/kg bw/day), respectively. The high contributions to PCB and 6 NDL-PCB intake corresponded to eel and mackerel, with approximately 40% of the total. The dietary DL-PCB intake was 7.27 pg-TEQ/day (0.12 pg-TEQ/kg bw/day), and the high contributions of DL-PCB intake were eel and mackerel, accounting collectively for approximately 45% of the total. The squid, anchovy, tuna, Spanish mackerel, and saury showed moderated contributions, accounting collectively for 30% of the total. This is associated with the relatively high consumption of this species as well as higher concentrations of PCBs in this species. In contrast, herring, showing the highest concentration, was the minor contributor due to the low consumption of herring, 0.1 g/day, in Korea.

Table 3
Estimated human exposure of PCBs and PBDEs by the general population of Korea, 2015–2017

	Total PCBs (ng/day)	6 NDL-PCBs[a] (ng/day)	12 DL-PCBs[b] (pg-TEQ/day)	Total PBDEs (ng/day)	7 PBDEs[c] (ng/day)
Eel	9.71	3.74	1.49	1.39	0.83
Mackerel	7.93	2.46	1.79	2.92	1.53
Squid	4.35	1.41	0.51	0.59	0.43
Anchovy	3.26	1.19	0.53	0.56	0.36
Tuna	2.98	1.06	0.38	0.64	0.42
Alaska pollack	2.38	0.83	0.18	0.17	0.12
Spanish mackerel	2.21	0.72	0.43	0.60	0.30
Hairtail	1.79	0.61	0.23	0.42	0.28
Rockfish	1.68	0.59	0.24	0.39	0.25
Saury	1.49	0.49	0.38	0.59	0.40
Flounder	1.15	0.43	0.16	0.25	0.16
Yellow croaker	0.91	0.27	0.08	0.24	0.14
Olive flounder	0.84	0.28	0.09	0.20	0.13
Herring	0.67	0.22	0.06	0.16	0.10
Cod	0.61	0.24	0.08	0.12	0.07
Mussel	0.48	0.18	0.15	0.12	0.09
Oyster	0.44	0.15	0.18	0.13	0.08
Octopus	0.36	0.13	0.04	0.06	0.04
Anger fish	0.32	0.12	0.02	0.05	0.03
Gizzard shad	0.27	0.09	0.02	0.04	0.02
Clam	0.26	0.09	0.02	0.05	0.04
Seabass	0.22	0.08	0.03	0.06	0.03
Crab	0.22	0.08	0.04	0.04	0.03
Filefish	0.14	0.05	0.00	0.03	0.02
Giant octopus	0.13	0.05	0.02	0.01	0.01
Shrimp	0.12	0.05	0.02	0.01	0.01
Puffer fish	0.10	0.03	0.01	0.02	0.01
Ark shell	0.07	0.02	0.04	0.01	0.01
Webfoot octopus	0.04	0.01	0.00	0.01	0.01
Abalone	0.03	0.01	0.00	0.01	0.01
Ray	0.02	0.01	0.00	0.00	0.00
Total (ng/day)	45.2	15.7	7.27	9.86	5.96
(ng/kg bw/day)[d]	0.76	0.26	0.12	0.17	0.10

a. Sum of 6 Non Dioxin-Like (NDL)-PCB congeners (CB-28, 52, 101, 138, 153, 180),

b. Sum of 12 DL-PCB congeners (CB-77, 81, 105, 114, 118, 123, 126, 156, 157, 167, 169 and 189),

c. Sum of 7 PBDE congeners (BDE-28, 47, 99, 100, 153, 154, and 183),

d. The body weight, 59.8 kg, was used to calculate the daily intakes for an average Korean.

For the risk assessment of PCBs through seafood consumption, we compared with recommendations, TDIs (tolerable daily intake) and TWI (tolerable weekly intake) established by Korea, Denmark, and WHO. The total PCB intake (0.76 ng/kg bw/day) was approximately 3.8% of the TDI (20 ng/kg bw/day) by WHO in 2003, and 0.8% of the TDI (100 ng/kg bw/day) by Danish Veterinary and Food Administration in 2012 (Carlsson et al. 2014). The DL-PCB intake (0.12 pg-TEQ/kg bw/day) was 3.0% of the TDI (4 pg-TEQ/kg bw/day) of dioxins (PCDD/Fs + DL-PCBs) by WHO and 6.0% of the TWI (Tolerable Weekly Intake, 14 pg-TEQ/kg bw/week) by Korea Food and Drug Administration (KFDA) (Moon and Choi 2009). These results suggest that the human exposure to the PCBs and DL-PCBs through seafood intake in Korea is safe. If the PCDD/F concentrations are added to the DL-PCB concentrations in view of their dioxin-like toxicity, the combined concentrations do not exceed the TDI by WHO and TWI by KFDA because contribution of dietary intake of PCDD/Fs (average 30% of DL-PCBs) was lower than DL-PCBs (Lee et al. 2007; Moon et al. 2009).

The total PBDE and 7 PBDE dietary intakes were 9.86 ng/day (0.17 ng/kg bw/day) and 5.96 ng/day (0.10 ng/kg bw/day), respectively. The high contributions to total PBDE and 7 PBDE intakes were also eel and mackerel, with approximately 40–45% of the total. The squid, tuna, Spanish mackerel, saury and anchovy also showed moderated contributions, accounting collectively for 30% of the total. To date, TDI levels have not been set for PBDEs by the EU due to limited data. A LOAEL (lowest observed adverse effect level) of 1 mg/kg/day has been reported for the most sensitive toxic effects of PBDEs (Martí-Cid et al. 2007). Benchmark doses per day (computed and estimated “safe levels” of the PBDEs) are currently 309 µg/kg bw/day for BDE-47, 12 µg/kg bw/day for BDE-99, and 83 µg/kg bw/day for BDE-153 (EFSA 2011). The European Food Safety Authority concluded that only BDE-99 would be of potential health concern for the European population (EFSA 2011). In this study, the dietary intake of 7 PBDEs including BDE-47, BDE-99 and BDE-153 (0.10 ng/kg bw/day) was less than 1% of the safe levels. According to the LOAEL and safe levels, the PBDE intake through seafood results in a safety factor of various orders of magnitude.

3.4. Temporal trends of concentrations of PCBs and PBDEs

To investigate the temporal trends of PCBs and PBDEs in seafood (the same 23 species), the concentrations, seafood consumptions, and dietary intakes of these contaminants in our study were compared to those reported in the previous surveys (MLTM 2012; Moon and Choi 2009). Data of concentrations, seafood consumption, and dietary intakes of DL-PCBs and 19 PBDEs for Korean general population are shown in Figs. 2 and 3.

The levels of DL-PCB in seafood were 0.05–3.20 pg-TEQ/g wet (mean: 0.87) in 2005–2007, 0.02–0.93 pg-TEQ/g wet (mean: 0.25) in 2010–2011, and 0.01–0.72 pg-TEQ/g wet (mean: 0.24) in 2015–2017. The decreasing rate during the 12-year period was 73% in average concentrations of DL-PCBs. The decreases of DL-PCB levels were observed in 22 of 23 species, and the declines in DL-PCB levels at more than 80%, were observed in six species (hairtail, mackerel, tuna, yellow croaker, crab, and shrimp). Kim and Yoon (2014) reported that average concentrations of dioxins/furans in air samples of Korea decreased 15 times from 0.79 pg-TEQ/m^3 in 1999 to 0.052 pg-TEQ/m^3 in 2009, and of DL-PCBs also decreased 10 times during the periods of 1999–2009. Jeong et al. (2016) also reported that levels of DL-PCBs in finless porpoises inhabiting Korean coastal waters showed significant decrease of 60–68% between 2003 and 2010 and regulations on POPs have been effective for marine mammals in Korea. Hence, these results indicated that reduction in PCB pollution might have occurred in Korea. A similar decreasing trends in PCB levels has recently reported in Indo-Pacific humpback dolphins (*Sousa chinensis*) collected from South China between 2004–2009 and 1995–2001 surveys, a representative biomonitor for contaminants in aquatic ecosystem (Wu et al. 2013). Sun et al. (2015) also reported that the PCB concentrations in two fish species from the Pearl River Estuary (South China) in 2013 were significantly lower than those in 2005 ($p < 0.05$), and declines of 61–80% were observed in two fish species during the 8-year period. This result indicates legislative actions on POPs such as PCBs have been effective in marine environments.

The levels of PBDEs in seafood were 0.02–1.55 ng/g wet (mean: 0.42) in 2010–2011, and 0.01–1.60 ng/g wet (mean: 0.35) in 2015–2017. The decreasing rate during the 7-year period was 17% in average concentrations of PBDEs. The decreases of DL-PCB levels were observed in 14 of 23 species, and the declines in PBDE levels at more than 50%, were observed in six species (anchovy, anger fish, mackerel, rockfish, crab, and shrimp). These results may reflect the ban on PBDEs and their decreased use due to the effectiveness of regulations and controls. In Korea, penta- and octa-BDE were banned in 2008 and usage of deca-BDE decreased 9 times during the periods of 2002–2010 (MOE 2015). Gauthier et al. (2008) determined PBDEs in herring gull eggs from the Laurentian Great Lakes (1982–2006). PBDE congeners derived mainly from penta-BDE and octa-BDE mixtures, BDE-47, -99, and − 100, showed rapid increases up until 2000, however, there are no increasing trend post-2000, due to regulation of penta- and octa-BDE mixtures. Since their phase outs in the 2000s, PBDE levels in herring gull eggs of 2012–2013 were 30% lower than in herring gull eggs of 2006–2008 (Su et al. 2015). The declining PBDE trend for edible portions of fish was observed in Great Lakes between 2006/07 and 2012 (Gandhi et al. 2017). The levels of PBDEs in environmental media expected to decline further since regulatory actions.

3.5. Temporal trends of human exposure

Total dietary intake of DL-PCB was 45.6 pg-TEQ/day in the 2005–2007 survey, 11.3 pg-TEQ/day in 2010–2011 and 6.18 pg-TEQ/day in 2015–2017 (Fig. 3). The decreasing rate during the 12-year period was 86% in total dietary intakes of DL-PCBs. The significant decreases of DL-PCB intakes between 2005–2007 and 2015–2017 surveys were found in four species, mackerel (15.8 pg-TEQ/day vs. 1.13 pg-TEQ/day), tuna (9.06 pg-TEQ/day vs. 0.55 pg-TEQ/day), hairtail (6.43 pg-TEQ/day vs. 0.34 pg-TEQ/day), and yellow croaker (3.64 pg-TEQ/day vs. 0.09 pg-TEQ/day), showing decreases of 93–98%. These species were consistent with those showing high declines in DL-PCB levels (hairtail, mackerel, tuna, yellow croaker,

crab, and shrimp). Moreover, the four species showed decrease of 45–68% in consumption. Therefore, the decrease of human exposure to DL-PCBs is mainly due to the decrease in concentrations of DL-PCB in seafood, along with seafood consumption among the surveys.

The estimated exposures of 19 PBDEs through fish and seafood consumption are shown in Fig. 3. Higher intake of PBDEs, 20.4 ng/day, was found in the 2010–2011 survey (MLTM 2012), while in the subsequent survey investigation, the estimated intake of PBDEs has been declining to 8.67 ng/day in the 2015–2017 survey. The decreasing rate during the 7-year period was 58% in dietary intakes of PBDEs. The significant decreases were found in anchovy (6.16 ng/day vs. 0.51 ng/day) and mackerel (6.05 ng/day vs. 1.87 ng/day), showing decreases of 69–92%. The decreasing rates of two species were 51–66% in PBDE concentrations and 37–75% in the consumption. The decrease of seafood intakes were found in 13 of 23 species and the decreasing rate was 27% in seafood consumption. Domingo et al. (2008) reported that the dietary intake of PBDEs through fish and shellfish intakes in Spain showed a decrease of 14% between 2000 and 2006, due to the decrease of 26% in fish and shellfish consumptions. Toms et al. (2018) analyzed serum pool concentrations of BDE 47, 99, 100 and 153 from 2002/03 to 2012/13, and reported that temporal trends were age- and congener-specific. Only two youngest age groups (0–4 and 5–15 year old) showed statistically significant decreases over the time, probably due to a decline in infant and toddler exposures in the indoor environment as use of PBDEs in consumer products has been phased out, however, older age groups showed no significant trend with time. These results indicate legislative actions on POPs have been effective in decrease of human exposure to POPs through seafood.

4. Conclusions

The exposures of Korean population to PCBs and PBDEs via seafood consumption have significantly decreased in 2015–2017, relative to those in 2005–2007 and 2010–2011. The significant decreases of PCB intakes were found in fatty fish such as mackerel, tuna, hairtail, yellow croaker, and anchovy. It is associated with the decrease in concentrations of PCBs and PBDEs in seafood between the surveys, due to regulatory actions. This indicates that levels and human exposure to PCBs and PBDEs from seafood consumption is expected to continue decreasing.

Declarations

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Figures

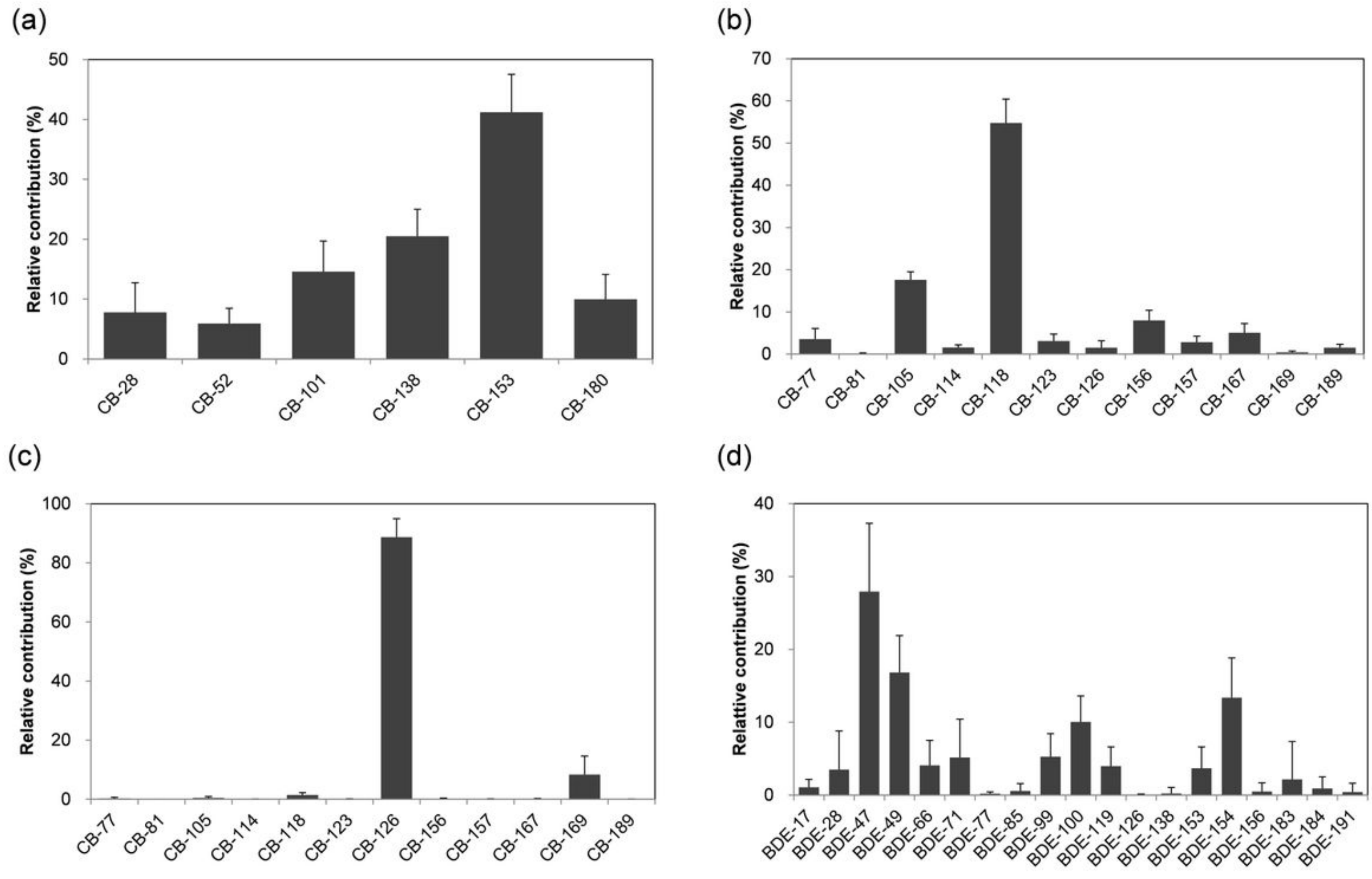

Figure 1

Relative contribution of (a) NDL-PCBs, (b) DL-PCBs (c) DL-PCBs (WHO-TEQ), and (d) PBDEs to their sum in seafood

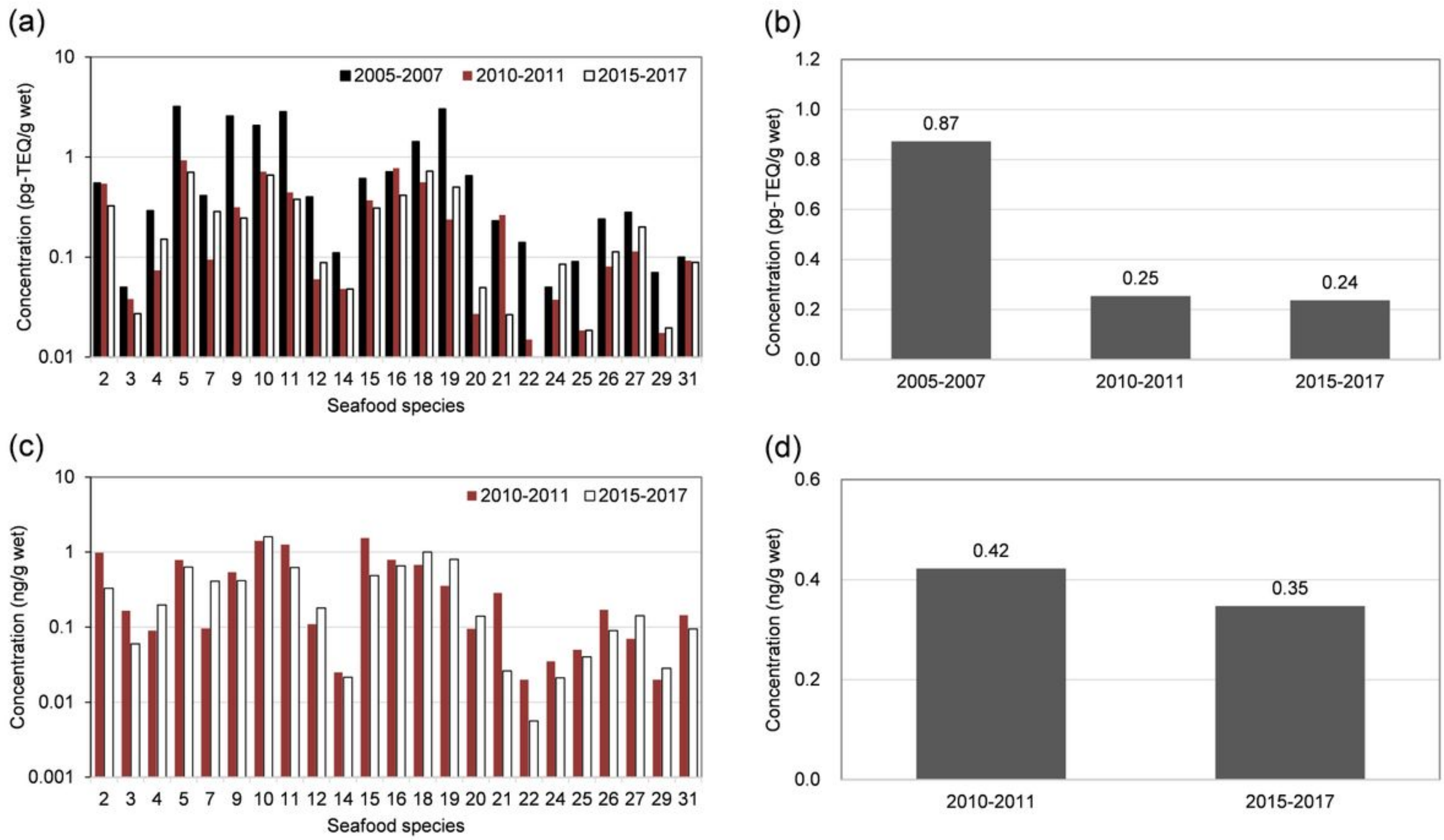

Figure 2

Temporal trend of (a) DL-PCB levels of each seafood species, (b) average DL-PCB levels of seafood, (c) PBDE levels of each seafood species, and (d) average PBDE levels of seafood in 2005-2017. The species names on the x-axes were presented in the same order as in Table 1

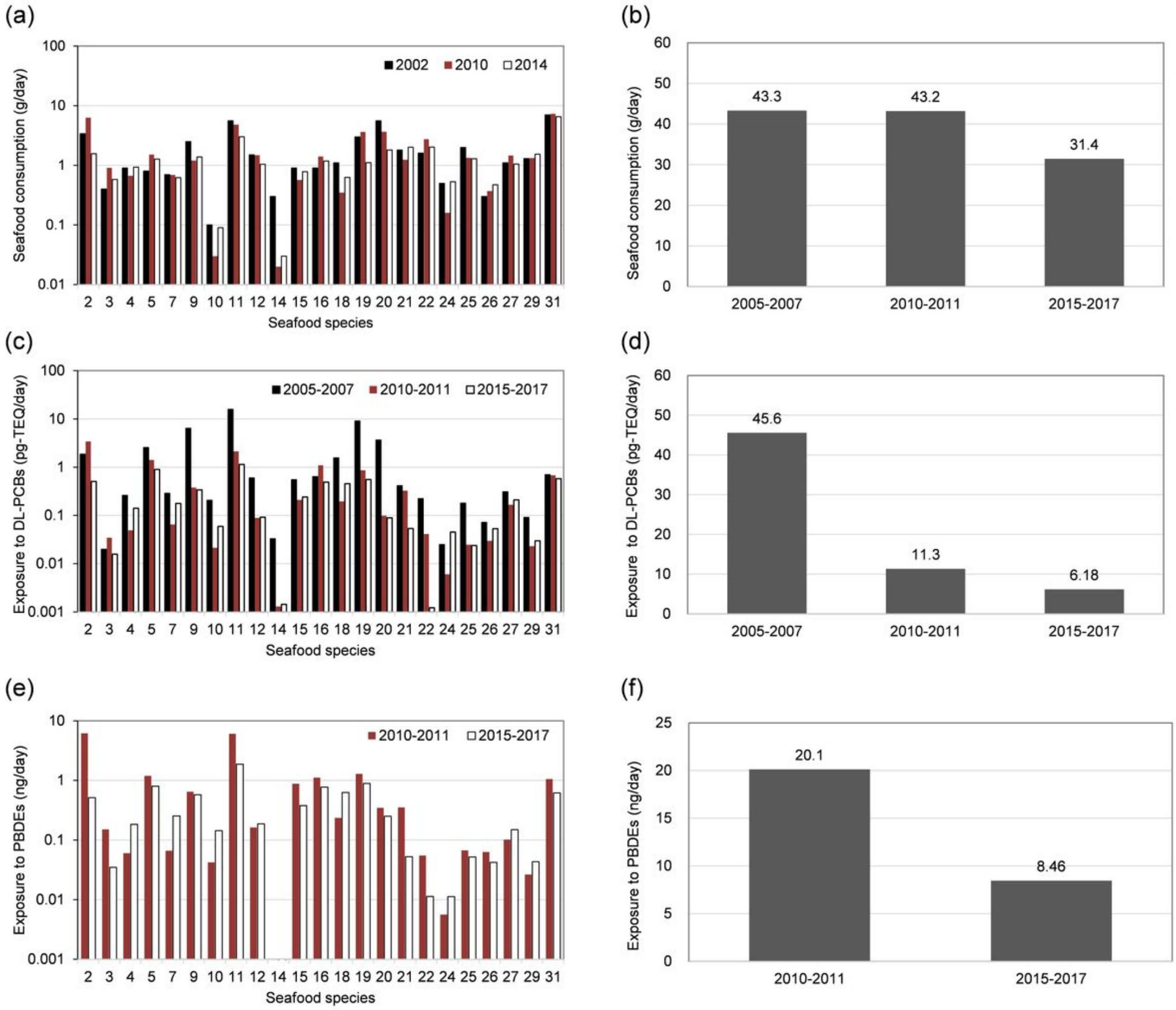

Figure 3

Temporal trend of (a) consumption for each seafood species and (b) total seafood consumption of Korean population, and dietary exposure of Korean population to DL-PCBs (c) by each seafood consumption and (d) by total seafood consumption, to PBDEs (e) by each seafood species and (f) by total seafood consumption. The species names on the x-axes were presented in the same order as in Table 1