



Review

Comparing levels of perfluorinated compounds in processed marine products

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ABSTRACT

Perfluorinated compounds (PFCs) are widely distributed in nature and have many applications due to their unique chemical and physicochemical properties. While, PFCs are present in soil, water, and air, their pathway for entry into the human body is circumstantially via contaminated food. The demand for seafood has been increasing. In this study, we investigated via LC-MS/MS, the content of 19 different types of PFCs in 302 samples belonging to five different categories of the typical South Korean seafood. The highest levels of PFOA, PFTrDA, PFOS, and PFPeA were found in dried seafood, canned and salted seafood, processed fish food, and seasoned laver, respectively. The levels of PFOA and PFOS were compared on the basis of various criteria including the nomenclature, biological classification, and habitat of the source of seafood. High levels of PFOA and PFOS were detected in anchovy, sea squirt, and mackerel based on the nomenclature of raw seafood, in crustaceans based on the biological classification, and in demersal organisms based on the habitat. The human intake values of PFOA and PFOS from the processed marine products in South Korea were lower than the tolerable daily intake, indicating that the consumption of these processed seafood poses no immediate harm.

1. Introduction

Anthropogenic contamination materials generated from industrial activities have been extensively studied (Sánchez-Avila et al., 2009). Among them, occurrences of perfluorinated compounds (PFCs) as persistent organic pollutants (POPs) in the environment have attracted widespread attention (Jensen and Leffers, 2008). PFCs are a class of organofluorine compounds characterized by a partially or fully fluorinated alkyl chain and a terminal functional group (carboxylate, sulfonate, sulfonamide, phosphonate, or alcohol). These compounds are non-flammable and resistant to chemical degradation (Ding and Peijnenburg, 2013). PFCs, due to their persistence, are found all over the world and have many industrial applications, including the manufacture of cleaner, paper coatings, and food packaging materials (Lau

et al., 2007; Paul et al., 2008).

The main exposure pathway of PFCs into the human body is circumstantially known to be via the intake of contaminated foods; PFCs are not easily excreted (Alexander et al., 2008; Kärrman et al., 2009; Domingo, 2012). Foods are possibly packaged in containers which may contain PFCs, giving cause for concern (Begley et al., 2005; Calafat et al., 2006; Tittlemier et al., 2007). Perfluorooctane sulfonate (PFOS) and perfluorooctanoate (PFOA), are the stable PFCs, are the main PFCs of concern owing to their toxicity and degree of dietary exposure (Kim et al., 2014), and hence, are the most studied among PFCs. PFOS and PFOA show moderate acute oral toxicity of LD₅₀ ~ 250 mg/kg b.w. (US EPA, 2000) and LD₅₀ ~ 680 mg/kg b.w. (Kennedy et al., 2004), respectively. Only in *in vitro* liver cells, PFOA and PFOS generate reactive oxygen species and oxidatively damage DNA (Eriksen et al., 2010). On

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the other hand, infants may be exposed to PFCs through breast milk, and PFOS intake may pose a potential risk for them (So et al., 2006). In addition, several PFCs were detected in human liver samples (Kärman et al., 2010).

PFCs are generally bound to proteins such as serum, and the half-lives of PFOA and PFOS in humans are 3.8 and 5.4 years, respectively (Olsen et al., 2005). Several studies reported the concentration of PFCs in human serum. The United States reported the mean serum values of PFOS (10.4–40.19 ng/mL), PFOA (2.08–6.98 ng/mL), and PFHxS (1.75–4.33 ng/mL) (Calafat et al., 2006a), South Korea reported median values of PFOS (7.96 ng/mL), PFOA (2.74 ng/mL), PFNA (2.09 ng/mL), PFUnDA (1.75 ng/mL), and PFHxS (1.51 ng/mL) (Ji et al., 2012), Spain reported the mean values of PFOS (7.64 ± 3.54 ng/mL), PFOA (1.80 ± 0.66 ng/mL), and PFHxS (3.56 ± 2.97 ng/mL) (Ericson et al., 2007), and Norway reported the mean value of PFOS (12 ng/mL), PFOA (2.7 ng/mL), and PFHxS (1.4 ng/mL) (Haug et al., 2009).

Of all the food categories, PFCs are most frequently detected in seafood (Falandysz et al., 2006). Recently, a number of studies reported the distribution of PFCs in water and marine organisms (Ericson et al., 2009; Jogsten et al., 2009; Zhang et al., 2011; Domingo et al., 2012; Vandermeersch et al., 2015). The global fish market is growing steadily, and reportedly had exports worth 140.0 US\$ billion in 2016 (<http://www.fao.org/in-action/globefish/fishery-information/resource-detail/en/c/338597/>). It is widely known that seafood is rich in nutrients, specially the ω -3 fatty acids, EPA and DHA, which may contribute to the prevention of heart diseases (Kris-Etherton et al., 2002). Consumption of a variety of seafood is recommended to meet the nutritional needs of children and pregnant woman (Dunstan et al., 2008).

In the present study, we monitored the levels of 19 PFCs in 302 processed seafood products as well as the South Korean products including dried seafood, salted seafood, and seasoned laver, which were obtained from the online and retail markets. Additionally, we analyzed the distribution pattern of 19 PFCs based on raw seafood nomenclature, crustacean on biological classification, and demersal organisms on habitat living in the near-bottom layer of the sea.

2. Materials and methods

2.1. Sampling

We obtained 302 food products from the online and retail markets and classified them into 2 categories, 'standard processed seafood' and 'other processed seafood'. We set the detailed categories and monitoring numbers on the basis of the data on "the sales and sale volume in Korea" (MFDS, 2014). The group of 'standard processed marine food' consisted of processed fish food (96), dried seafood (29), and salted seafood (43). Canned food (105) and seasoned laver (29) were categorized under 'other processed seafood'. The detailed information of samples is summarized in Table 1.

2.2. Homogenization and sample storage

The purchase dates of all the samples were within 1 year of each other. The samples were stored at -20°C before being opened. All the samples were blended with water (HPLC grade) using a glass food processor. The homogenized ratio of samples is summarized in Table 1. All the homogenized samples were stored in a polypropylene conical tube at -20°C .

2.3. Chemicals and materials

HPLC-grade water, hexane, acetonitrile (ACN), and *tert*-butyl methyl ether (MTBE) were purchased from Burdick & Jackson (B&J, Ulsan, South Korea). Sodium carbonate ($\geq 99.5\%$), sodium bicarbonate ($\geq 99.7\%$), and tetrabutylammonium hydrogensulfate (97%) were purchased from Sigma-Aldrich (St. Louis, MO, USA). Protease from

Streptomyces griseus and lipase from *Candida rugose* were purchased from Sigma-Aldrich (St. Louis, MO, USA). Ammonium hydroxide ($\geq 25\%$ in H_2O , for LC-MS) and ammonium acetate ($\geq 99.0\%$, for LC-MS) were purchased from Sigma Aldrich. A mixture of native perfluoroalkylcarboxylates and native perfluoroalkylsulfonates, perfluoro-*n*-butanoic acid (PFBA, 98%), -pentanoic acid (PFPeA, 98%), -hexanoic acid (PFHxA, 98%), -heptanoic acid (PFHpA, 98%), -octanoic acid (PFOA, 98%), -nonanoic acid (PFNA, 98%), -decanoic acid (PFDA, 98%), -undecanoic acid (PFUnDA, 98%), -dodecanoic acid (PFDoA, 98%), -tridecanoic acid (PFTrDA, 98%), -tetradecanoic acid (PFTeDA, 98%), potassium perfluoro-1-butanedisulfonate (PFBS, 98%), sodium perfluoro-1-hexanesulfonate (PFHxS, 98%), sodium perfluoro-1-heptanesulfonate ($> 98\%$, PFHpS), sodium perfluoro-1-octanesulfonate (PFOS, 98%), and sodium perfluoro-1-decanedisulfonate (PFDS, 98%) was purchased from Wellington Laboratories (Guelph, Ontario, Canada). Perfluoro-1-octanesulfonamide ($> 98\%$, PFOA), *N*-methylperfluoro-1-octanesulfonamidoacetic acid ($> 98\%$, MeFOSAA), and *N*-ethylperfluoro-1-octanesulfonamidoacetic acid ($> 98\%$, EtFOSAA) were purchased from Wellington Laboratories (Guelph, Ontario, Canada). The mixture containing nine isotopically labelled reference standards, [$^{13}\text{C}_2$]PFHxA (98%), [$^{13}\text{C}_4$]PFOA (98%), [$^{13}\text{C}_5$]PFNA (98%), [$^{13}\text{C}_2$]PFDA (98%), [$^{13}\text{C}_2$]PFUnDA (98%), [$^{13}\text{C}_2$]PFDoDA (98%), [$^{18}\text{O}_2$]PFHxS (98%), [$^{13}\text{C}_4$]PFOS (98%), and [$^{13}\text{C}_4$]PFBA (98%) was purchased from Wellington Laboratories (Guelph, Ontario, Canada). Single stable-isotope-labelled *N*-methyl- d_3 -perfluoro-1-octanesulfonamidoacetic acid (d_3 -NMeFOSAA, 98%), *N*-ethyl- d_5 -perfluoro-1-octanesulfonamidoacetic acid (d_5 -NEtFOSAA, 98%), [$^{13}\text{C}_5$]PFPeA (98%), [$^{13}\text{C}_4$]PFHpA (98%), and [$^{13}\text{C}_2$]PFTeDA (98%), which were used as internal standards were purchased from Wellington Laboratories (Guelph, Ontario, Canada).

2.4. Extraction and analysis

Lipase (7000 units/mL) and protease (30 units/mL) were added to the homogenized samples (1 g) after spiking 20 μL of the internal standard (IS) mixture that comprised of 100 ng/mL of the nine isotopically labelled reference standards and 50 $\mu\text{g}/\text{mL}$ each of the stable-isotope-labelled standards mentioned in section 2.3, and then the samples were incubated for 16 h. The enzyme-treated samples were shaken with 5 mL of hexane, and the supernatant was removed. This process was then repeated. In the ion-pairing method, the samples were sonicated for 10 min after adding 1 mL of 0.5 M tetrabutylammonium hydrogensulfate and 2 mL of a mixture of 0.25 M sodium carbonate and sodium bicarbonate. For the liquid-liquid extraction, 5 mL of *tert*-butyl methyl ether was added and shaken for 30 min. After shaking and separating the solid and liquid phases using a centrifuge, the supernatant was transferred into a conical tube and was evaporated to dryness under nitrogen on a sample concentrator. It was then eluted with 200 μL of ACN. The final step involved filtration using a 0.2- μm nylon filter with a polypropylene housing. We applied the same extraction method to dried seafood, canned food, processed fish food, and salted seafood. For seasoned laver, a modified method was applied. The flow chart of the extraction method is shown in Supplementary Fig. 1.

Analysis of samples was performed using liquid chromatography coupled to a tandem mass spectrometer (LC-MS/MS) with the electrospray interface operating in the negative ion mode. Separation was carried out using a YMC-ODS C18 column (2.0 mm \times 150 mm, 3 μm). The mobile phase gradient was operated at a flow rate of 0.2 mL/min. The starting eluent contained 5 mM ammonium acetate dissolved in 0.02% aqueous formic acid (A, 70%) and MeOH (B, 30%). At the middle of the analysis time, MeOH content was increased to 100%. 30% (B) was flown from 13 min to the end of the analysis time. The final analysis was performed using a multiple reaction monitoring method (Table 2).

Table 1
Summarized information of samples and the concentrations of PFOA, PFOS and sum of the other PFCs.

Category	Subcategory	PFOA (ng/g)	PFOS (ng/g)	Σ17 PFCs (ng/g)	Raw material	Biological classification	Country of origin	Habitat	Packaging type	Homogenized ratio (sample:water)
Dried seafood (29)	Dried food (4)	mean min-max	0.24 < LOQ, < LOQ, 0.26–0.54	0.09 < LOQ, < LOQ, 0.06–0.56	Bonito (1), pollack (3)	Fish (4)	Indonesia (1), Russia (3)	Pelagic (4)	Polyethylene	1:8
	Seasoned dried food (17)	mean min-max	0.4 < LOQ, < LOQ, 0.11–0.44	0.16 < LOQ, < LOQ, 0.04–1.25	Squid (14), filefish (2), pollack (1)	Cephalopod (14), Fish (3)	Peru (11), Vietnam (2), Russia (1), Chile (1), domestic (2)	Pelagic (15), Demersal (2)	Polyethylene, Polypropylene	1:3
	Other dried food (8)	mean min-max	0.09–1.74 1.52 0.15–3.00	1.07 < LOQ, < LOQ, 0.19–4.25	Dried mussel (1), shrimp (1), anchovy (1), squid (2), seaweed (2), cod(1)	Cephalopod (2), Fish (2), Seaweed (2), Shellfish (1), Crustacean (1)	China (1), Vietnam (1), domestic (6)	Pelagic (3), Demersal (5)	Polyethylene	1:2 (1), 1:3 (4), 1:7 (2), 1:8 (1)
	Mackerel canfood (10)	mean min-max	0.08 < LOQ, 0.04–0.31	0.77 0.52–1.14	Mackerel (10)	Fish (10)	Domestic (10)	Pelagic (10)	Tin can	1:1
Canned seafood (105)	Whelk canfood (11)	mean min-max	0.43 < LOQ, 0.05–0.62	0.16 < LOQ, 0.01–1.84	Whelk (11)	Shellfish (11)	Imported (8), Mexico (1), Bulgaria(1), domestic (1)	Demersal (11)	Tin can	1:1
	Oyster Canfood (4)	mean min-max	0.13 < LOQ, 0.04–0.29	0.12 < LOQ, 0.00–0.62	Oyster (4)	Shellfish (4)	Domestic (4)	Demersal (4)	Tin can	1:1
	Pacific saury canfood (10)	mean min-max	0.05 < LOQ, 0.07–0.12	0.11 < LOQ, 0.02–0.83	Pacific saury (10)	Fish (10)	Imported (10)	Pelagic (10)	Tin can	1:1
	Salmon canfood (20)	mean min-max	0.05 < LOQ, 0.03–0.07	0.08 < LOQ, 0.17	Salmon (20)	Fish (20)	The US (17), Chile (3)	Pelagic (20)	Tin can	1:1
Processed fish food (96)	Tuna canfood (50)	mean min-max	0.05 < LOQ, 0.04–0.09	0.08 < LOQ, 0.01–0.82	Tuna (50)	Fish (50)	Imported (50)	Pelagic (50)	Tin can	1:1
	Fish cake (73)	mean min-max	0.05 < LOQ, 0.04–0.09	0.06 < LOQ, 0.02–0.95	Mixed fish fillet (62), pollack (7), golden-thread (3), croaker (1)	Fish (11), Mixed fish fillet (62)	Imported (55), the US (8), Vietnam (7), Pakistan (1), Indonesia (1), domestic (1)	^a All (62), Pelagic (8), Demersal (3)	Polyethylene	1:2
	Fish Sausage (12)	mean min-max	0.05 < LOQ	0.08 < LOQ, 0.02–2.14	Mixed fish fillet (8), pollack (4)	Mixed fish fillet (12)	Imported (9), The US (3)	^a All (8), Pelagic (4)	Polyvinylidene chloride	1:2
	Paste (2)	mean min-max	0.1 < LOQ, 0.16	0.95 0.61–1.29	Mixed fish fillet (2)	Mixed fish fillet (2)	China (1), Pakistan (1)	^a All (2)	Polyethylene	1:2
Other processed seafood (9)	Other processed seafood (9)	mean min-max	0.08 < LOQ, 0.05–1.34	0.128 < LOQ, 0.01–1.22	Mixed fish fillet (8), eel (1)	Mixed fish fillet (8), Fish (1)	Imported (5), Vietnam (2), domestic (2)	^a All (8), Demersal (1)	Polyethylene, Polypropylene	1:1 (1), 1:3 (6), 1:4 (1), 1:5 (1)

(continued on next page)

Table 1 (continued)

Category	Subcategory	mean	min-max	PFOA (ng/g)	PFOS (ng/g)	Σ17 PFCs (ng/g)	Raw material	Biological classification	Country of origin	Habitat	Packaging type	Homogenized ratio (sample:water)	
Collective salted seafood (43)	Fermented seafood (2)	mean	0.16	0.19	0.1	< LOQ,	Halibut (2)	Fish (2)	Domestic (2)	Demersal (2)	Polyethylene	1:1	
		min-max	0.16–0.17	0.18–0.20	0.01–0.56	< LOQ,							
	Fish sauce (7)	mean	0.05	0.08	0.05	< LOQ,	Anchovy (4), sand lance (3)	Fish (7)	Domestic (7)	Domestic (7)	Pelagic (7)	Polyethylene terephthalate	1:1
		min-max	< LOQ	< LOQ	< LOQ,	< LOQ,							
Seasoned fish sauce (6)	mean	0.04	0.07	0.05	0.01–0.09	Anchovy (5), sand lance (1)	Fish (6)	Thailand (3), domestic (3)	Thailand (3), domestic (3)	Pelagic (6)	Polyethylene terephthalate	1:1	
	min-max	< LOQ,	< LOQ,	< LOQ,	< LOQ,								
Salted seafood (8)	mean	0.61	1.09	0.14	0.03–0.10	Shrimp (4), anchovy (2), yellow corvina (1), clam (1)	Crustacean (4), Fish (3), Shellfish (1)	Domestic (8)	Domestic (8)	Pelagic (3), Demersal (5)	Polyethylene	1:1	
	min-max	0.06–2.64	0.18–2.26	0.01–1.56	0.18–2.26								
Seasoned salted seafood (20)	mean	0.55	0.36	0.15	0.02–0.07	Squid (4), pollack roe (5), small octopus (2), sea squirt (2), oyster (2), anchovy (1), hairtail (1), dried cuttlefish (1), pollack (1)	Cephalopod (7), Fish (9), Shellfish (2), Other (2)	Domestic (12), Russia (5), China (2), the US (1)	Domestic (12), Russia (5), China (2), the US (1)	Pelagic (12), Demersal (8)	Polyethylene	1:1	
	min-max	< LOQ,	< LOQ,	< LOQ,	0.02–3.80								
Seasoned laver (29)	mean	0.39	0.42	0.15	0.08–2.01	Seaweed (29)	Seaweed (29)	Domestic (29)	Domestic (29)	Demersal (29)	Polyethylene, Polyethylene terephthalate	1:20	
	min-max	< LOQ,	< LOQ,	< LOQ,	0.46–2.62								

Numbers in parentheses indicate sample size.

min-max: minimum-maximum.

^a “All” is the mixed fish fillet (uncertain fish origin) including pelagic fish and (or) demersal fish.

Table 2
Mass transitions (parent ion/product ion) for the analyzed PFCs and method validation followed via linearity, LOQ, and LOQ.

Compound	Abbreviation	Primary ion transition (m/z)	Secondary ion transition (m/z)	LOD	LOQ	r ²
Perfluoro-n-butanoic acid	PFBA	212.903 → 168.8	212.903 → 168.2	0.06	0.18	0.9997
Perfluoro-n-pentanoic acid	PFPeA	262.98 → 218.9		0.04	0.12	1.0000
Perfluoro-n-hexanoic acid	PFHxA	313 → 268.9	313 → 119	0.03	0.10	0.9999
Perfluoro-n-heptanoic acid	PFHpA	363.105 → 319	363.105 → 168.7	0.04	0.11	0.9999
Perfluoro-n-octanoic acid	PFOA	413 → 368.7	413 → 169.1	0.03	0.09	1.0000
Perfluoro-n-nonanoic acid	PFNA	462.98 → 418.9	462.98 → 218.9	0.03	0.08	0.9999
Perfluoro-n-decanoic acid	PFDA	512.91 → 468.8	512.91 → 218.9	0.06	0.18	0.9999
Perfluoro-n-undecanoic acid	PFUnDA	562.83 → 519	562.83 → 268.9	0.02	0.06	0.9996
Perfluoro-n-dodecanoic acid	PFDoDA	612.96 → 569	612.96 → 168.7	0.01	0.04	0.9999
Perfluoro-n-tridecanoic acid	PFTTrDA	662.994 → 618.8	662.994 → 168.9	0.02	0.07	0.9999
Perfluoro-n-tetradecanoic acid	PFTeDA	713.1 → 669.1	713.1 → 168.9	0.03	0.10	1.0000
Potassium perfluoro-1-butanedisulfonate	PFBS	298.981 → 79.8	298.981 → 98.9	0.02	0.06	0.9998
Sodium perfluoro-1-hexanedisulfonate	PFHxS	399 → 79.9	399 → 98.8	0.02	0.07	1.0000
Sodium perfluoro-1-heptanedisulfonate	PFHpS	449.038 → 98.9	449.038 → 168.9	0.01	0.04	0.9998
Sodium perfluoro-1-octanedisulfonate	PFOS	498.964 → 80	498.964 → 98.9	0.05	0.15	0.9998
Sodium perfluoro-1-decanedisulfonate	PFDS	598.889 → 80	598.889 → 99.1	0.03	0.10	0.9999
Perfluoro-1-octanesulfonamide	PFOA	498.937 → 79.8	498.937 → 98.9	0.03	0.09	1.0000
N-methylperfluoro-1-octanesulfonamidoacetic acid	MePFOSAA	569.896 → 419.1	569.896 → 482.7	0.06	0.18	0.9999
N-ethylperfluoro-1-octanesulfonamidoacetic acid	EtPFOSAA	583.969 → 418.7	583.969 → 526	0.03	0.09	0.9997
Perfluoro-n-[1,2–13C2]hexanoic acid	[13C2]PFHxA	315.1 → 269.8				
Perfluoro-n-[1,2,3,4–13C4]octanoic acid	[13C4]PFOA	417.1 → 371.9				
Perfluoro-n-[1,2,3,4,5–13C5]nonanoic acid	[13C5]PFNA	468 → 423				
Perfluoro-n-[1,2–13C2]decanoic acid	[13C2]PFDA	514.9 → 469.9				
Perfluoro-n-[1,2–13C2]undecanoic acid	[13C2]PFUnDA	564.9 → 519.9				
Perfluoro-n-[1,2–13C2]dodecanoic acid	[13C2]PFDoDA	614.9 → 570				
Sodium perfluoro-1-hexane[18O2]sulfonate	[18O2]PFHxS	403.02 → 83.9	403.02 → 102.9			
Sodium perfluoro-1-[1,2,3,4–13C4]octanedisulfonate	[13C4]PFOS	503 → 80	503 → 99			
Perfluoro-n-[13C4]butanoic acid	[13C4]PFBA	216.919 → 172				
N-methyl-d3-perfluoro-1-octanesulfonamidoacetic acid	d3-NMeFOSAA	573 → 418.9				
N-ethyl-d5-perfluoro-1-Octanesulfonamidoacetic acid	d5-NEFOSAA	588.9 → 419.1	588.9 → 531.1			
Perfluoro-n-[13C5]pentanoic acid	[13C5]PFPeA	269.93 → 118.9				
Perfluoro-n-[1,2,3,4–13C4]heptanoic acid	[13C4]PFHpA	367.105 → 322				
Perfluoro-n-[1,2–13C2]tetradecanoic acid	[13C2]PFTeDA	715.121 → 670				

2.5. Validation and quality control

The external standard curve with 8 points in the range of 0–10 ng/g with the IS mixture diluted in ACN was obtained. To validate the methods used to measure the contamination in various seafood products, we applied the same extraction method to each group. The limit of detection (LOD) and the limit of quantification (LOQ) were calculated using the y-intercepts' standard deviation of regression lines (A) and the slope of the calibration curve (B): $LOD = 3.3(A)/(B)$; $LOQ = 10(A)/(B)$. We performed intra-day (n = 7) and inter-day (n = 21) validation to confirm the accuracy and precision of the method. We spiked the 20 µL of standard solution representing low (0.5 ng/g), medium (1 ng/g), and high (5 ng/g) concentrations to food mixture samples, to determine recovery. The accuracy range and precision range were between 80 and 120% and within 20%, respectively. Additionally, we performed quality control with the standard solutions of concentrations 0.2 and 0.5 ng/g, and repeatedly checked the instrument sensitivity after every 20 samples.

2.6. Statistical analysis

For the comparison of the levels of PFOA and PFOS in different groups of food products, we tested testing difference between the means using a censored regression (p -value < 0.05). Values less than LOQ were replaced by the half value of the LOQ for statistical analysis, and its natural logarithm was used.

3. Results and discussion

3.1. Validation and quality control

We validated our extraction and analysis methods. Linearity (R^2) of the calibration curve of PFCs was more than 0.9996, LOD ranged

between 0.01 and 0.06 ng/g and LOQ ranged from 0.04 to 0.18 ng/g (Table 2). Depending on the matrix or extraction method for measuring PFCs, we performed the validation with four food product groups; processed seafood (dried seafood and processed fish food), canned seafood, collective salted seafood, and seasoned laver (Supplementary Tables 1 and 2). The average intra-day recovery representing the accuracy (n = 7) ranged from 80 to 120% for three concentrations (0.5, 1, and 5 ng/g). Further, relative standard deviation, which represents the precision, was within 20%. Validation was performed three times on different days to determine the inter-day accuracy and precision. Accuracy ranged between 80 and 120%, while precision was within 20%. We developed an extraction method for seasoned lavers with increased sonication time and an additional boiling procedure. Additionally, we performed cross-analysis with Eulji University (Seongnam, South Korea) using the same extraction method. The accuracy of the cross analysis was within 20%.

3.2. Concentrations of 19 PFC species in classified seafood samples

A few studies have reported the levels of PFCs in processed seafood. PFC levels in seafood and their products were monitored in China (Wu et al., 2012), Hong Kong (Zhao et al., 2011), the United Kingdom (Clarke et al., 2010), and Canada (Tittlemier et al., 2007). However, to the best of our knowledge, a comparative analysis of the PFC levels in various types of processed seafood products, including salted food, dried seafood, and seasoned laver, has not been conducted. Additionally, while PFOA and PFOS are the main PFCs reported in literature, data for other types of PFCs is limited. Therefore, we conducted this study to monitor the levels and detection rates of all the 19 PFC species in five different food groups (Table 3). We summarized the concentrations of PFOA, PFOS, and the sum of the concentrations of other 17 PFCs in subcategories with the sample information (Table 1). In canned food, the only PFC with a detection

Table 3
Concentrations (ng/g) of 19 PFCs in the processed marine products.

Dried seafood (29)	PFBA	PFPeA	PFHxA	PFHpA	PFOA	PFNA	PFDA	PFUnDA	PFDoDA	PFTrDA
	< LOQ	16	10	7	9	5	15	23	9	12
average	0.18 ± 0.14	0.26 ± 0.21	0.26 ± 0.25	0.27 ± 0.26	0.69 ± 0.83	0.20 ± 0.40	0.15 ± 0.12	0.36 ± 0.40	0.11 ± 0.12	0.34 ± 0.61
min	0.16	0.09	0.05	0.06	0.09	0.05	0.25	0.06	0.04	0.06
median	0.31	0.34	0.27	0.32	0.50	0.18	0.30	0.43	0.16	0.28
max	0.68	0.82	1.14	1.18	3.00	2.07	0.52	1.25	0.42	2.53
	PFTeDA	PFBS	PFHxS	PFHpS	PFOA	PFDS	PFOSA	MePFOSAA	EtPFOSAA	
< LOQ	19	21	5	21	10	11	23	29	29	
average	0.12 ± 0.14	0.07 ± 0.08	0.39 ± 0.29	0.04 ± 0.05	0.44 ± 0.82	0.10 ± 0.07	0.08 ± 0.08	0.09 ± 0.00	0.05 ± 0.00	
min	0.08	0.04	0.06	0.04	0.11	0.07	0.12	0.00	0.00	
median	0.25	0.13	0.48	0.06	0.28	0.13	0.19	0.00	0.00	
max	0.49	0.40	1.09	0.28	4.25	0.35	0.34	0.00	0.00	
Canned food (105)	PFBA	PFPeA	PFHxA	PFHpA	PFOA	PFNA	PFDA	PFUnDA	PFDoDA	PFTrDA
< LOQ	23	39	74	80	48	35	84	16	21	10
average	0.20 ± 0.15	0.11 ± 0.12	0.06 ± 0.06	0.07 ± 0.05	0.07 ± 0.07	0.06 ± 0.04	0.10 ± 0.03	0.20 ± 0.33	0.06 ± 0.08	0.26 ± 0.51
min	0.08	0.04	0.04	0.04	0.03	0.03	0.07	0.03	0.02	0.03
median	0.19	0.10	0.06	0.07	0.06	0.06	0.10	0.09	0.03	0.07
max	0.81	0.82	0.45	0.44	0.62	0.26	0.20	1.55	0.55	2.65
	PFTeDA	PFBS	PFHxS	PFHpS	PFOA	PFDS	PFOSA	MePFOSAA	EtPFOSAA	
< LOQ	54	71	71	41	38	74	30	82	93	
average	0.08 ± 0.06	0.04 ± 0.03	0.11 ± 0.22	0.03 ± 0.02	0.20 ± 0.28	0.09 ± 0.11	0.14 ± 0.19	0.10 ± 0.04	0.05 ± 0.02	
min	0.04	0.03	0.03	0.01	0.06	0.05	0.04	0.06	0.04	
median	0.08	0.03	0.11	0.03	0.12	0.12	0.09	0.13	0.08	
max	0.41	0.19	1.40	0.18	1.93	0.99	0.92	0.27	0.14	
Processed fish food (96)	PFBA	PFPeA	PFHxA	PFHpA	PFOA	PFNA	PFDA	PFUnDA	PFDoDA	PFTrDA
< LOQ	70	72	89	92	82	81	95	48	48	59
average	0.13 ± 0.14	0.12 ± 0.28	0.05 ± 0.03	0.06 ± 0.04	0.05 ± 0.03	0.05 ± 0.02	0.08 ± 0.02	0.09 ± 0.10	0.04 ± 0.04	0.06 ± 0.08
min	0.05	0.03	0.03	0.02	0.04	0.02	0.15	0.03	0.01	0.03
median	0.15	0.10	0.06	0.22	0.07	0.08	0.15	0.12	0.05	0.07
max	1.22	2.14	0.36	0.35	0.26	0.13	0.15	0.66	0.30	0.53
	PFTeDA	PFBS	PFHxS	PFHpS	PFOA	PFDS	PFOSA	MePFOSAA	EtPFOSAA	
< LOQ	76	91	81	77	15	94	40	95	94	
average	0.06 ± 0.03	0.03 ± 0.01	0.04 ± 0.03	0.02 ± 0.02	0.29 ± 0.27	0.05 ± 0.01	0.16 ± 0.22	0.08 ± 0.02	0.04 ± 0.02	
min	0.05	0.01	0.02	0.01	0.05	0.06	0.05	0.15	0.06	
median	0.09	0.05	0.07	0.03	0.20	0.07	0.15	0.15	0.12	
max	0.16	0.06	0.19	0.12	1.34	0.08	1.06	0.15	0.19	
Collective salted seafood (43)	PFBA	PFPeA	PFHxA	PFHpA	PFOA	PFNA	PFDA	PFUnDA	PFDoDA	PFTrDA
< LOQ	28	36	32	40	14	18	33	18	19	14
average	0.11 ± 0.06	0.07 ± 0.03	0.05 ± 0.02	0.06 ± 0.02	0.39 ± 0.95	0.13 ± 0.21	0.10 ± 0.02	0.30 ± 0.41	0.07 ± 0.09	0.43 ± 0.53
min	0.04	0.04	0.03	0.05	0.02	0.03	0.08	0.01	0.00	0.07
median	0.14	0.06	0.04	0.06	0.16	0.12	0.12	0.36	0.08	0.53
max	0.35	0.22	0.17	0.22	4.57	1.08	0.19	1.56	0.42	2.58
	PFTeDA	PFBS	PFHxS	PFHpS	PFOA	PFDS	PFOSA	MePFOSAA	EtPFOSAA	
< LOQ	26	36	11	29	15	33	25	43	39	
average	0.07 ± 0.04	0.05 ± 0.10	0.17 ± 0.60	0.02 ± 0.01	0.40 ± 0.56	0.06 ± 0.03	0.14 ± 0.19	0.09 ± 0.00	0.05 ± 0.01	
min	0.04	0.03	0.01	0.01	0.03	0.02	0.03	0.00	0.02	
median	0.08	0.06	0.04	0.02	0.28	0.05	0.16	0.00	0.04	
max	0.24	0.56	3.80	0.05	2.26	0.26	0.71	0.00	0.07	

(continued on next page)

Table 3 (continued)

Seasoned laver (29)	PFBA	PFPeA	PFHxA	PFHpA	PFOA	PFNA	PFDA	PFUnDA	PFDoDA	PFTrDA
< LOQ	29	23	27	29	10	29	29	14	11	20
average	0.09 ± 0.00	0.56 ± 1.81	0.08 ± 0.14	0.05 ± 0.00	0.39 ± 0.30	0.04 ± 0.00	0.09 ± 0.00	0.24 ± 0.24	0.26 ± 0.29	0.20 ± 0.28
min	0.00	0.00	0.43	0.00	0.38	0.00	0.00	0.26	0.18	0.35
median	0.00	1.28	0.56	0.00	0.49	0.00	0.00	0.37	0.37	0.45
max	0.00	9.59	0.69	0.00	1.11	0.00	0.00	0.79	1.22	1.14
	PFTeDA	PFBS	PFHxS	PFHpS	PFOS	PFDS	PFOSA	MePFOSAA	EtPFOSAA	
< LOQ	26	21	28	9	19	29	29	29	29	
average	0.13 ± 0.26	0.18 ± 0.29	0.05 ± 0.10	0.24 ± 0.18	0.42 ± 0.60	0.05 ± 0.00	0.04 ± 0.00	0.09 ± 0.00	0.05 ± 0.00	
min	0.49	0.30	0.56	0.18	0.46	0.00	0.00	0.00	0.00	
median	0.75	0.57	0.56	0.34	0.91	0.00	0.00	0.00	0.00	
max	1.23	1.03	0.56	0.63	2.62	0.00	0.00	0.00	0.00	

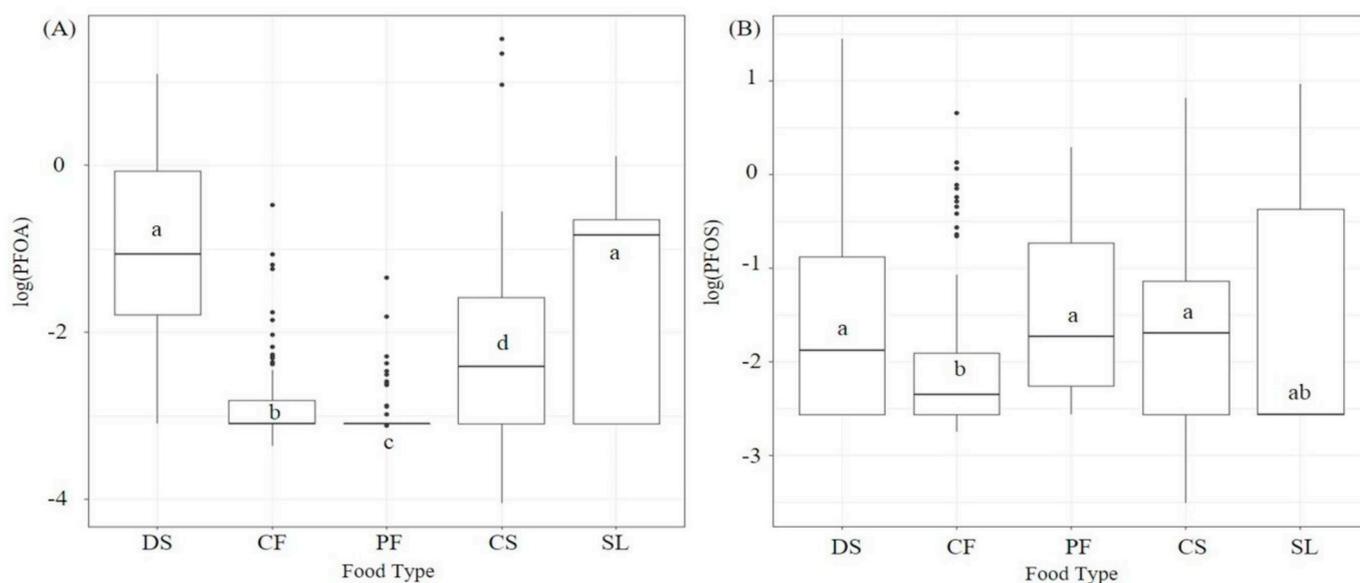


Fig. 1. Box plots of (A) perfluorooctanoate (PFOA) and (B) perfluorooctane sulfonate (PFOS) in processed marine products by classifying processing methods. DS = dried seafood, CF = canned food, PF = processed fish food, CS = collective salted seafood, SL = seasoned laver (line = median, box = 25th and 75th percentiles, whiskers = minimum and maximum observations below upper fence) \times IQR (Inter Quantile Range) = 75th percentile – 25th percentile. Means within box plots with different letters are significantly different at $p < 0.05$ by testing differences between means using a censored regression.

rate higher than 90% was PFTrDA (90.5%). PFCs with the highest detection rates below the LOQ (< LOQ) were MePFOSAA (100%) and EtPFOSAA (100%) in dried seafood, EtPFOSAA (88.6%) in canned food, PFDA (99.0%) and MePFOSAA (99.0%) in processed fish food, MePFOSAA (100%) in collective salted seafood, and PFBA (100%), PFHpA (100%), PFNA (100%), PFDA (100%), PFDS (100%), PFOSA (100%), MePFOSAA (100%), and EtPFOSAA (100%) in seasoned laver (Table 3). The species of PFCs detected at the highest average concentrations were PFOA (0.69 ± 0.83 ng/g) in dried seafood, PFTrDA (0.26 ± 0.51 ng/g) in canned food, PFOS (0.29 ± 0.27 ng/g) in processed fish food, PFTrDA (0.43 ± 0.53 ng/g) in collective salted seafood, and PFPeA (0.56 ± 1.81 ng/g) in seasoned laver (Table 3). In dried seafood, the average concentration ranged from < LOQ to 0.69 ng/g, and PFOS (4.25 ng/g) was detected at the highest concentration (max value) (Table 3). Additionally, PFOS was detected at the max value in whelk canned food (1.93 ng/g), paste (1.29 ng/g), other processed seafood (1.34 ng/g), and seasoned fish sauce (0.10 ng/g), and the PFOA was detected at the max value in salted seafood (2.64 ng/g), and seasoned salted seafood (4.57 ng/g) (Table 1). The levels of PFOA and PFOS were generally higher than the levels of other PFCs in most of the food categories. But, some of the other 17 PFCs were detected at the max value in the category of canned food (except whelk canfood), fish cake, fish sausage,

fermented seafood, fish sauce, and seasoned laver (Table 1). While the contents of PFOA, PFOS, and PFOSA ranged from 1.2 ng/g to 5.1 ng/g, 0.7–12.8 ng/g, and 1.1 and 1.7 ng/g, respectively in a Czech study (Hradkova et al., 2010), the PFCs detected at high levels in canned food in this study were PFTrDA (0.26 ± 0.51 ng/g), PFUnDA (0.20 ± 0.33 ng/g), PFBA (0.20 ± 0.15 ng/g), and PFOS (0.20 ± 0.28 ng/g) (Table 3). The total average PFC level (5.47 ng/g) in canned mackerel was higher than that in canned whelk, canned oyster, canned pacific saury, canned salmon, and canned tuna. The PFC species detected at the highest concentration in canned mackerel was PFTrDA, followed by PFUnDA (Supplementary Table 3). Similarly, a PFUnA (the same with PFUnDA) concentration of 3.1 ± 0.212 ng/g was reported in mackerel in Portugal (Alves et al., 2017). The PFCs detected at the highest concentrations in their study were PFBA in salmon and tuna, PFPeA in oyster, PFHxS in whelk, and PFUnDA in Pacific saury (Alves et al., 2017). In processed fish food, the concentrations of all the PFC species were below 0.10 ng/g, except for PFBA, PFPeA, PFOS and PFOSA with the average concentrations of 0.13, 0.12, 0.29, and 0.16 ng/g, respectively. Further, in collective salted seafood, PFC species occurred at concentrations below 0.17 ng/g, except for PFOA, PFUnDA, PFTrDA, PFHxS, and PFOS with the concentrations of 0.39, 0.30, 0.43, 0.17, and 0.40 ng/g respectively, while in seasoned laver, the average concentrations of PFCs more than

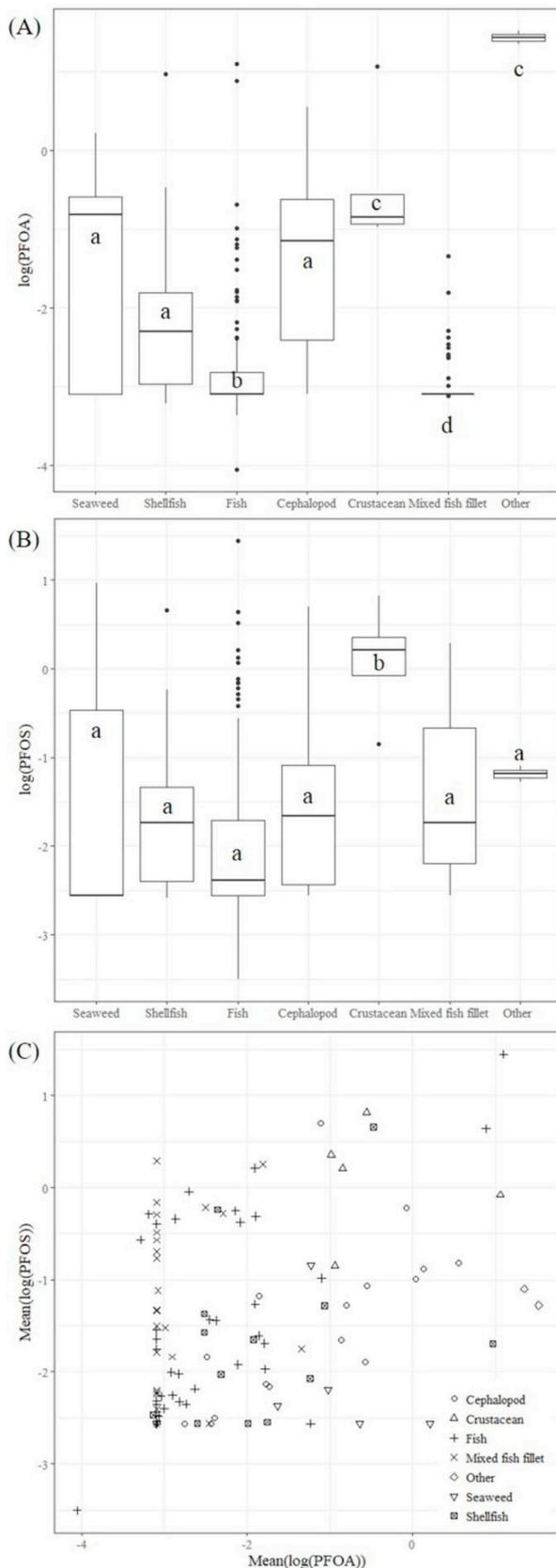


Fig. 2. Box plots of PFOA (A) and PFOS (B) in all processed marine products based on biological classification: seaweed, shellfish, fish, cephalopod, crustacean, mixed fish fillet, and other (sea squirt). (C) is the distribution of a sample's concentration to compare PFOA and PFOS. (line = median, box = 25th and 75th percentiles, whiskers = minimum and maximum observations below upper fence) \approx IQR (Inter Quantile Range) = 75th percentile – 25th percentile. Means within box plots with different letters are significantly different at $p < 0.05$ by testing differences between means using a censored regression.

3.3. Comparing the concentration of PFOA and PFOS in samples

The serum levels of PFOA and PFOS are known to be higher than those of other PFCs (Kim et al., 2014). Fromme and Tittlemier, 2009 reviewed that the diet contributed to 96% and 99% of the direct PFOS and PFOA intake, respectively. In the present study, the median concentration of PFOA in dried seafood was significantly higher ($P < 0.05$) than that in other food groups except seasoned laver which is a dried type of food (Fig. 1). The high levels of PFCs in dried seafood were mainly attributed to the high PFC levels in anchovies (pelagic), squids (pelagic), cods (demersal), and shrimps (demersal); the concentration of PFOA was higher than that of PFOS (Supplementary Table 4). In a previous study, the median concentration of PFOA was 21.2 ng/g and that of PFOS was below the LOD in squids, and the median concentrations of PFOA and PFOS were 20.6 ng/g, 0.19 ng/g, respectively, in shrimps (Chang et al., 2012). However, some studies showed that the concentrations of PFOS were higher than those of PFOA in anchovies and cods. PFOA and PFOS concentrations in haddock, cods, and soles in Canada were less than 0.5 ng/g and 2.6 ng/g, respectively (Tittlemier et al., 2007). The concentration of PFOA in raw anchovies was less than the LOD, while that of PFOS was 3.06 ± 0.10 ng/g in Greece (Vassiliadou et al., 2015). In our study, the highest PFOS concentration in anchovies was 4.25 ng/g (Supplementary Table 4). The similarity in PFOS concentration could be due to the comparable PFOS level in raw anchovies. Because PFOS is a stronger acid than PFOA, the bioaccumulation rate of PFOS is higher (Ding and Peijnenburg, 2013). On the other hand, the concentration of PFOS in only canned food was significantly lower than that of dried seafood, processed fish food, and collective salted seafood except seasoned laver (Fig. 1). Although the levels of PFCs in the processed seafood products, but in the raw marine materials, were monitored in this study, these differences in the PFC levels in salted or dried samples compared to the other processed or canned products could be partly due to the water-loss during drying or salting. Vassiliadou et al. (2015) reported that the water-loss by evaporation or oil-uptake could increase the concentration of PFCs in the cooked samples as a function of the mass loss.

To investigate the differences by categories, we compared the detailed results, which are illustrated in Supplementary Fig. 2. The levels of PFOS were broadly distributed in different categories of processed seafood compared with those of PFOA. The concentrations of PFOA in the subcategory of canned food were not significantly different. Although each canned food contained the same materials, the PFOS concentrations in the subcategory of canned food were different. The levels of PFOS in oysters, and pacific sauries were significantly different with those of mackerel, whelk, and tuna (Supplementary Fig. 2). The mean concentration of PFOS (0.654 ± 0.240 ng/g) in blue fish (Ericson et al., 2008) was similar to that in canned mackerel 0.77 ± 0.22 ng/g, which is a kind of blue fish, analyzed in our study (Supplementary Table 3). Further, a previous study reported that tuna, sardines, and mussels had PFOS levels of 0.271 ± 0.029 ng/g and PFOA levels less than 0.126 ng/g (Ericson et al., 2008). Tuna, cod, and plaice, which are lean fishes, had PFOS and PFOA concentrations of 308 ng/g and 23 ng/g, respectively (Noorlander et al., 2011).

On the other hand, we performed statistical analyses for investigating whether the concentrations of PFOA and PFOS were

0.20 ng/g were PFPeA (0.56 ng/g), PFOA (0.39 ng/g), PFUnDA (0.24 ng/g), PFDoDA (0.26 ng/g), L-PFHpS (0.24 ng/g), and PFOS (0.42 ng/g) (Table 3).

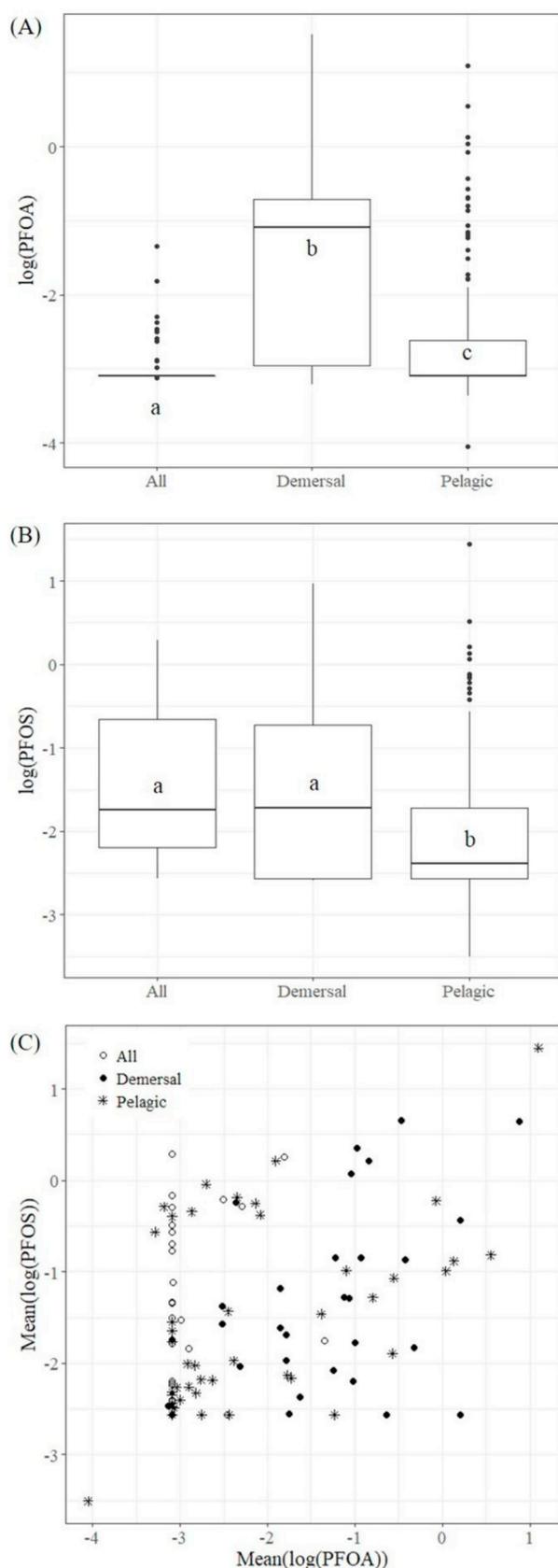


Fig. 3. Box plots of PFOA (A) and PFOS (B) to compare demersal and pelagic living organisms. (C) is the distribution of a sample's concentration to compare PFOA and PFOS. 'All' is the mixed fish fillet (uncertain), the organisms that have untraceable habitats (line = median, box = 25th and 75th percentiles, whiskers = minimum and maximum observations below upper fence). \approx IQR (Inter Quantile Range) = 75th percentile – 25th percentile. Means within box plots with different letters are significantly different at $p < 0.05$ by testing differences between means using a censored regression.

(Fig. 2). This category (“Other”) consists of salted sea squirt. Sea squirts mainly inhabit the sea bottom or are attached to the rocks. The high levels of PFOS and PFOA were detected in shell fishes such as whelks and clams that share the same habitat as sea squirts (Quinete et al., 2009). Furthermore, our data showed that the partial marine organisms that inhabit the bottom of the sea, such as seaweeds (e.g., laver and kelp) and crustaceans, had higher levels of PFOA and PFOS than the fish that mainly live in water far from bottom (Fig. 2).

Based on these results, we analyzed the samples depending on whether the sources of the samples inhabit the pelagic or demersal habitats (Fig. 3). We organized the reported levels of PFCs in samples from several countries based on their pelagic and demersal origins (Table 4). The findings of studies that monitored the samples from the demersal locations, mussels and oysters (Munschy et al., 2013) and mussels in Brazil (Quinete et al., 2009), samples from pelagic locations (various fish samples (Berger et al., 2009)), and the samples from both pelagic and demersal locations (Barbarossa et al., 2016), are organized in Table 4. A study that compared pelagic and benthonic fishes (demersal) reported the PFOS concentrations of 4 ng/g in pelagic fishes and 13 ng/g in benthonic fishes and the PFOA concentrations of 3 ng/g in pelagic fishes and 14 ng/g in benthonic fishes (Nania et al., 2009). In other countries, the level of PFOA was 46 pg/g and that of PFOS was 582 pg/g in crustaceans whereas the level of PFOA was 23 pg/g and that of PFOS was 308 pg/g in lean fishes (e.g., cod, plaice, pollack, and tuna) (Noorlander et al., 2011). The range of PFC concentrations (mg/kg) in crabs (demersal) was 11–20, sardines (pelagic) was 1–7, sprats (pelagic) was 3–8, and eels (both) was 51–63 (Clarke et al., 2010). The range of PFOS concentrations in fishes (e.g., small yellow croaker and silvery pomfret) was 0.38–2.93 ng/g and crabs was 0.94–4.59 ng/g. The concentration of PFOA in fishes was below the LOQ while that in crabs was 0.34–1.67 ng/g (Gulkowska et al., 2006) (Table 4).

Furthermore, in the present study, laver collected from the bottom of the sea floor or from the coast showed relatively high levels of PFOS and PFOA (Supplementary Fig. 2). We concluded that the concentrations of PFOS and PFOA in demersal fish were higher than those in pelagic fish based on the results in Table 4 and of this study. Due to environmental contamination, demersal organisms are exposed to higher levels of PFCs in water, sludge, rocks, and sand than pelagic organisms (Zareitalabad et al., 2013). Another concern is that only the demersal organisms had higher PFOA levels than the PFOS levels and had significantly higher PFOA levels ($P < 0.05$) than those in the categories “All” and pelagic organisms (Fig. 3). This could be explained by the fact that concentrations of PFOA are higher than those of PFOS in the sea deposits (Nakata et al., 2006).

4. Conclusion

We showed that the levels of PFCs, including PFOA and PFOS, were highly different in various processed marine products consumed in Korea and abroad through monitoring. The concentrations of PFTrDA, and PFPeA as well as PFOA and PFOS were higher than those of other PFCs. We consider that the PFCs in marine products mainly originate from the raw materials. Furthermore, we performed statistical analyses based on the differences in the biological classifications and habitats of the source of the food. The highest levels of PFOA and PFOS were detected in crustaceans. Furthermore, we found that the organisms which

affected by the biological classification in our samples. The seafood category of “Other” had the highest PFOA levels and a significantly higher level than that in other food categories except “crustacean”

Table 4
Overview of recent studies on PFC concentrations in fish (raw) and seafood materials.

Country	Characteristics of the study	PFCs analyzed	Sample	Samples habitat	Results of analysis	Reference
China	PFCs levels in 92 seafood from coastal area	13 PFCs including PFOA and PFOS	Turbot, belt fish, yellow croaker, red sea bream, oyster scallop, clam, red drum and so on	Pelagic and demersal	Range PFOA pg/g: Fatty fish; 16–8500 and Shellfish; 94–7970 PFOS pg/g: Fatty fish; 63–8500 and Shellfish; 8- < 2000	(Wu et al., 2012)
Hong Kong	Analysis of PFCs in freshwater fish and marine fish	10 PFCs including PFOA and PFOS	Freshwater fish (10) and marine fish (10)	Pelagic and demersal	PFOA mg/g: freshwater fish; 1.2–1.4 and marine fish; 1.0 PFOS ng/g: freshwater fish; 0.27–5.98 and marine fish; 0.27–2.39	(Zhang et al., 2011)
United Kingdom	PFCs levels measured in UK retail foods	11 PFCs including PFOA and PFOS	13 fish species and 5 shellfish crustaceans	Pelagic and demersal	Range PFCs mg/kg: Crab; 11–20, Whitebait; 8–62, Eel. 51–63, Sardines; 1–7, Sprats; 3–8	(Clarke et al., 2010)
Canada	PFCs levels measured in fish, and their packaging	6 PFCs including PFOA and PFOS	Canned fish (salmon, tuna), shrimp, trout, cod and so on	Pelagic and demersal	PFOA < 0.5 ng/g PFOS < 2.6 ng/g	(Tittlemier et al., 2007)
Czech	PFCs contamination in canned fish and seafood products	PFOA, PFOS, and PFOSA	Canned fish (sardine, cod, herring, mackerel, mussel, octopod, tuna, sprat, and so on)	Pelagic and demersal	The highest value µg/kg of PFOA (5.1; sardine), PFOS (12.8; sardine), and PFOSA (1.7; sardine)	(Hradkova et al., 2010)
Portugal	Contaminants of raw and cooked seafood	PFOS, PFUnA	Mackerel, and flounder	Pelagic	Total PFCs concentration ng/g (flounder and mackerel): raw; 24.0 and 3.1, steamed; 22.0 and 2.9	(Alves et al., 2017)
France	Levels of PFCs in mussels and oysters	PFOS	Mussels and Oyster	Demersal	Mussels and Oyster: 0.005–0.9 ng/g	(Munsch et al., 2013)
Netherlands	Concentration of PFCs at food and dietary intake	14 PFCs including PFOA and PFOS	9 fish species, 3 crustaceans, and so on	Pelagic and demersal	PFOS (pg/g d.w.): lean fish (308), crustaceans (582), fatty fish (61)	(Noorlander et al., 2011)
China	Determination of PFCs in food and drinking water	10 PFCs including PFOA and PFOS	Salmon, shrimp, oyster, clam, and squid	Pelagic and demersal	The highest value ng/g: PFOA; oyster (34.7), PFDA; oyster (22.2)	(Chang et al., 2012)
Greece	Levels of PFCs in raw and cooked finfish and shellfish	12 PFCs including PFOA and PFOS	Finfish (anchovy, bogue, hake, picarel, sardine, sand smelt and striped mullet), mussel, shrimp and squid	Pelagic and demersal	Raw, fried, grilled samples Anchovy: PFUnDA (1.50, 1.75, and 2.73), PFDoA (1.86, 2.99, and 3.52), Picarel: PFOS (20.37, 44.69, and 1.16) and so on.	(Vassiliadou et al., 2015)
Spain	Levels of PFCs in food from market	11 PFCs including PFOA and PFOS	White fish, seafood, tinned fish, and blue	Pelagic	PFOA: < 0.029- < 0.132 ng/g PFOS: 0.148–0.654 ng/g	(Ericson et al., 2008)
Brazil	Specific profiles of accumulation in biological samples	10 PFCs including PFOA and PFOS	Mussels from various area in Brazil	Demersal	PFOS: < 0.95–4.46 ng/g PFOA: < 0.84–6.02 ng/g	(Quinete et al., 2009)
Sweden	Analysis of edible fish from Lake Vättern and the Baltic Sea	11 PFCs including PFOA and PFOS	Perch, burbot, whitefish, salmon, and brown trout	Pelagic	PFOS was the highest PFCs in all samples from salmon 0.98 and 8.49, perch 11.3 and 2.13, burbot 12.0 and 1.69, whitefish 2.86 and 2.51, and brown trout 5.73 and 1.08 (ng/g)	(Berger et al., 2009)
Italy	Analysis of PFCs concentration from fish that was consumed in Italy	PFOA and PFOS	5 fish species (sea bass, mullet, hake, plaice, and mackerel)	Pelagic and demersal	Range of PFOS (ng/kg): sea bass (703–1243), mullet (487–1896), hake (91–1292), plaice (240–510), mackerel (54–120)	(Barbarossa et al., 2016)
Italy	Monitoring of PFCs in edible fish from the Mediterranean Sea	PFOS and PFOA	Pelagic fishes (muscle and liver), benthonic fishes (muscle and liver), squids, redshanks, mussels and clams	Pelagic and demersal	Range of PFOA (ng/kg): sea bass (93–487), mullet (12–113), hake (traces-127), plaice (10–36), mackerel (traces-22) Mean of PFOS (ng/g): pelagic fishes (muscle 4, liver 13), benthonic fishes (muscle 13, liver 53), cephalopod molluscs (squids 3, redshanks 3), bivalve molluscs (mussels < 2, clams 2)	(Nania et al., 2009)
China	Levels of PFCs in china from two locations	8 PFCs including PFOA and PFOS	6 fish species, 2 mollusc, 2 crabs, 3 shrimps, and 4 shellfishes	Pelagic and demersal	Mean of PFOA (ng/g): pelagic fishes (muscle 3, liver 6), benthonic fishes (muscle 14, liver 9), cephalopod molluscs (squids 2.5, redshanks 2.5), bivalve molluscs (mussels < 1.5, clams 15) Range of PFOS (ng/g): fish (0.38–2.93), mollusc (0.87–1.32), crab (0.94–4.59), shrimp (0.58–1.3.9), and shellfish (0.42–0.54) Range of PFOA (ng/g): fish (< LOQ), mollusc (0.31–0.43), crab (0.34–1.67), shrimp (0.42–0.45), shellfish (0.27–0.48)	(Gulkowska et al., 2006)

live close to the sea floor, had higher PFOA and PFOS levels. Among the 302 samples that we analyzed, the max values of PFOA and PFOS were obtained from sea squirt (4.57 ng/g) and anchovy (4.25 ng/g), respectively. The daily average intake of sea squirt and anchovy was 384 g (95 percentile) and 8.06 g (100 percentile), respectively, by Korean (Korea, 2012). Based on these data, the consumption of PFOA and PFOS was 25 ng/kg/d and 0.49 ng/kg/d, respectively assuming a 70 kg average body weight for an adult. The intake value of PFOA and PFOS from the processed marine products in South Korea were lower than the tolerable daily intake (TDI) for PFOA (1500 ng/kg/d), and PFOS (150.0 ng/kg/d), set by EFSA (Alexander et al., 2008), indicating no likely immediate harm with respect to the current dietary intake of PFCs via the processed seafood products in South Korea.

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Transparency document

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Appendix A. Supplementary data

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