J. Fish. Sci. Tech. 5(2), 127~135, 2002

JOURNAL OF
FISHERIES SCIENCE
AND TECHNOLOGY

Accumulation and Characterization of Polycyclic Aromatic Hydrocarbons in Seafood from the Coastal Areas of Korea

Hyo-Bang Moon*, Hee-Gu Choi, Sang-Soo Kim and Pil-Yong Lee

Marine Environment Management Division, National Fisheries Research & Development Institute, Busan 619-902, Korea

(Received February 2002, Accepted June 2002)

Twenty seafood samples, which are common edible species and commercially important items in Korea, were purchased at the local fisheries markets and were analyzed for polycyclic aromatic hydrocarbons (PAHs) using gas chromatography coupled to mass spectrometer detector (GC/MSD). The levels of sixteen PAHs in seafood from Korean coasts were 161 to 2,243 pg/g wet weight. The highest concentration was found at saury (*Coloabis saira*) and the lowest level was found at jacopever (*Sebastes schlegeli*). The concentrations of potentially carcinogenic PAHs of six species were in the range of 9 to 123 pg/g wet weight. The residues of PAHs in fishes from Korean coasts were slightly low or relatively moderate to other countries. There was no correlation between PAH residues and lipid contents in seafood samples. The predominant contributors in fish samples were lower-molecular-weight two and three ring aromatic PAHs such as naphthalene, acenaphthene, fluorene and phenanthrene. Filter-feeding organisms like shrimp, crab and topshell were dominated by three- and four-ring aromatic PAHs. The PAH profiles in marine sediments, bivalves, fishes, shrimp, crab and topshell according to exposure pathway were compared through factor analysis. The PAH profiles were clearly classified by the difference of species or environmental matrices. This result suggests that most of PAHs within the same samples behave identically in marine environment.

Key words: Seafood, Polycyclic aromatic hydrocarbons (PAHs), GC/MSD, Residues, Factor analysis

Introduction

Polycyclic aromatic hydrocarbons (PAHs) are ubiquitous environmental microcontaminants and derive mainly from anthropogenic activities. They are formed as a consequence of incomplete combustion (Hites et al., 1977) and are also components of crude oil and its refined products (Lee and Page, 1997; Pettersen et al., 1997). Since they are carcinogenic and mutagenic (Lehr and Jerina, 1977; Yan, 1985; Mix. 1986; White, 1986), these compounds have been intensively studied in marine environment (Wakeham et al., 1980; Prahl and Carpenter, 1983; Law and Biscaya, 1994; Baumard et al., 1999; Soclo et al., 2000).

Marine organisms may be exposed to PAH by contact with contaminated seawater and sediments, either on the seabed or through suspended sediments, or by ingestion of contaminated prey (Hellou et al., 1996). The different PAH profiles of contaminants have been observed in organisms of different trophic levels (Broman et al., 1990; Leonards et al., 1997). These differences were attributed to a partial biotransformation of the contaminants in the organisms of higher trophic levels (Baumard et al., 1998). Therefore, these organisms reflect the pollution extent of PAHs and some species are used as bioindicators at different environmental conditions and foodweb (Hellou and Warren, 1996; Escartn and Porte, 1999).

PAHs are known to occur in aquatic environments and accumulate in aquatic organisms (Adamo et al., 1997; Hendriks et al., 1998; Burkhard and

*Corresponding author: hbmoon@nfrdi.re.kr

Lukasewycz, 2000), so that consumption of fish and topshell may be a significant human dietary source for these chemicals eventually posing a real health risk (Fairey et al., 1997; Sericano et al., 2001). Hence, the determinations of these compounds in seafood are very important for evaluating dietary exposure assessment and the protection of public health, particularly in view of the increasing availability to the consumer of seafood.

Although PAHs have previously been determined in some locations for sediments and bivalves from Korean coastal environments (Lee et al., 1998; Kim et al., 1999; Moon et al., 2001a; Moon et al., 2001b), there are few reported data on levels of PAHs in seafood including fish from Korean coasts. The main purpose of the current study was to measure the PAH residues in edible tissue of seafood species most often caught from Korean coasts and to investigate bioaccumulation patterns of PAHs in different organism species.

Materials and Methods

Sample collection and preparation

Twenty seafood samples were purchased at local fisheries markets from nine locations distributed over Korean coastal areas (two from the eastern region, five from the southern region, and two from the western region) during July to August 2001. The samples of sixteen fish species, common squid (*Todarodes pacificus*) from East Sea, crab (*Portunus trituberculatus*), shrimp (*Penaeus orientails*) from West Sea, topshell (*Batillus cornutus*) from South Sea were collected in this investigation. These seafood species are common edible species and are commercially important food items in Korea. Biological information on the seafood species is summarized in Table 1.

The samples in a cooler box with ice or dry ice were immediately transported to the laboratory. The seafood tissues of eighteen specimens and the whole bodies of two specimens (anchovy and whitesaddled reeffish) were homogenized with an ultradisperser. Homogenized samples (approximately 50 g) were decomposed in 200 mL of 1 N KOH ethanolic solution for 2 hours by mechanical shaking after the spike of 7 species internal standards (ES 2044, Cambridge Isotope Laboratories, Inc.). The digest was liquid-liquid extracted with twice using 150 mL of *n*-hexane (Ultra residue analysis, J.T. Baker) after the addition of water and 50 g of anhydrous Na_2SO_4. The extracts were reduced to small volume in a rotary evaporator and then adjusted to a volume of 10 mL.

Table 1. Biological characteristics of studied seafood samples

Local market	Species	Sample code	Length (cm)	Height (cm)
East Sea				
Gangleung	Roundnose flounder (*Eopsetta grigorjewi*)	RF	20	8
Gangleung	Common squid (*Todarodes pacificus*)	CS	28	15
Pohang	Saury (*Cololabis saira*)	SU	28	3
Pohang	Herring (*Clupea pallasii*)	HR	27	6
South Sea				
Busan	Hairtail (*Trichiurus lepturus*)	HT	31	8
Busan	Mackerel (*Scomber japonicus*)	MK	42	6
Tongyeong	Anchovy (*Engraulis japonica*)	AN	4	1
Tongyeong	Common conger (*Conger myriaster*)	CC	32	3
Yeosu	Sharp toothed eel (*Muraenesox cinereus*)	STE	33	4
Yeosu	Red tongue sole (*Cynolossus joyneri*)	RTS	14	9
Yeosu	Sea bass (*Lateolabrax japinicus*)	SB	38	11
Yeosu	Silver fish (*Pampus argenteus*)	SF	19	10
Mokpo	Flatfish (*Paralichthys olivaceus*)	FF	43	16
Mokpo	Jacopever (*Sebastes schlegeli*)	JP	30	10
Jeju	Topshell (*Batillus cornutus*)	TS	7	4
Jeju	Whitesaddled reeffish (*Chromis notata*)	WR	5	3
West Sea				
Gunsan	Korean shrimp (*Penaeus orientails*)	KS	14	2.5
Gunsan	Korean pomfret (*Pampus echinogaster*)	KP	20	10
Incheon	Greening (*Hexagrammos otakii*)	GR	25	5
Incheon	Blue crab (*Portunus trituberculatus*)	BC	18	10

The extracts of seafood were purified using an activated silica gel (Art No. 7734, 70~230 mesh, Merck) column chromatography with successive elutants of *n*-hexane and 15% methylene dichloride (Pesticide residue analysis, Cica-Merck) in *n*-hexane. The second fraction was concentrated to less than 1 mL, and left at a room temperature for one day to evaporate to 100~200 μL. The residues were dissolved with 100 μL of *n*-nonane (Pesticide residue analysis, Fluka) and determined for PAHs.

Quantification and identification

The aromatic fraction was analyzed by gas chromatography coupled to mass spectrometery. An Agilent 6890 GC (Agilent, USA) equipped with a split/splitless injector was used (splitless time: 2 min; flow 70 mL/min). The injector temperature was maintained at 250℃. The GC temperature program was from 80℃ (1 min) to 300℃ (10 min) at 4℃/min. The carrier gas was helium at a constant flow rate of 1.4 mL/min. The capillary column used was a PTE-5 (30 m, 0.32 mm ID, 1.0 μm film thickness, Supelco). The GC was coupled to a 5973N mass selective detector (MSD). The mass spectrometer was operated under the selected ion monitoring (SIM) mode using molecular ions of the investigated PAHs (electron impact at 70 eV, 2000 V, 1.5 scan/s, dwell time/ion: 40 ms). The interface temperature was 250℃.

For lipid determination, automatic extraction unit (Gerhardt and Variostat, Germany) was used. About 2 g of freeze-dried seafood sample was extracted with 150 mL of *n*-hexane. The analytical condition was as follows; boiling time: 30 min, solvent reduction A: 5×15 mL, extraction time: 80 min, solvent reduction B: 8 min, solvent reduction C: 5 min, solvent reduction interval: 4 min. Extracted samples were evaporated to dryness at ambient temperature and the lipid fraction was weighed.

Sixteen non-alkylated PAH compounds recommended by US EPA were analyzed in each sample (naphthalene (NaP), acenaphthylene (AcPy), acenaphthene (AcP), fluorene (Flu), phenanthrene (PhA), anthracene (AnT), fluoranthene (FluA), pyrene (Pyr), benzo[a]anthracene (BaA), chrysene (Chr), benzo[b]fluoranthene (BbF), benzo[k]fluoranthene (BkF), benzo[a]pyrene (BaP), indeno[1,2,3-c,d]pyrene (InP), dibenzo[a,h]anthracene (DbA) and benzo[g,h,i]perylene (BghiP)).

Quality assurance

All the spiked internal standards were detected with no interfering peak. The average recoveries showed 71±10% for two, 80±7% for three, 84±5% for four, 88±3% for five, and 94±9% for six ring aromatic groups. In order to assess the accuracy of the determinations by experimental procedure and instrument, the certified mussel homogenate (1974a, NIST, USA) was analyzed as Standard Reference Materials (SRMs) in this investigation. The recovery results for two-, three-, four-, five-, and six-ring aromatic groups were 68%, 77%, 87%, 94% and 107%, respectively. Procedural blanks were processed in the same manner as real samples, and they were below 10% of analytes abundance. Blanks were run before and after the injection of standard solutions to check for any carryover. The calculated detection limit (S/N ratio=3) for individual PAH in seafood sample was 1 pg/g wet weight.

Results and Discussion

PAHs concentrations in seafood

Sixteen PAHs were detected in all seafood samples from Korean coastal areas. The PAHs contents in seafood are summarized in Table 2. The lipid contents (dry weight basis) in seafood were in the range of 1.2~48.3%, showing a wide range. The levels of ΣPAH (the sum of individual PAH) in seafood from Korean coast were 161 to 2,243 pg/g wet weight. The highest concentration was found at saury (*Cololabis saira*) from East Sea and the lowest level was found at jacopever (*Sebastes schlegeli*) from South Sea. Total concentrations of potentially carcinogenic PAHs (ΣCPAH; the sum of benzo[a]anthracene, benzo[b]fluoranthene, benzo[k]fluoranthene, benzo[a]pyrene, indeno[1,2,3-c,d]pyrene, and dibenzo[a,h]anthracene) (IARC, 1987) in seafood varied from 9 to 123 pg/g wet weight. The flatfish (*Paralichthys olivaceus*) from South Sea was characterized by the highest contents of ΣCPAH. In particular, the highest level of the benzo[a]pyrene, which was known as one of endocrine disruptor chemicals, was found at this species and the proportion of this compound to total PAH

Table 2. The PAHs contents in seafood from Korean coastal areas (pg/g wet weight)

	RF	CS	SU	HR	HT	MK	AN	CC	STE	RTS	SB	SF	FF	JP	TS	WR	KS	KP	GR	BC
NaP	232	53	40	41	210	16	81	32	25	30	14	25	32	25	53	56	54	27	36	23
AcPy	37	51	12	17	14	26	80	34	9	9	18	9	9	7	17	18	14	9	8	9
AcP	59	363	1,726	1,003	1,328	773	1,407	456	191	38	203	194	55	38	82	1,563	75	81	32	40
Flu	116	89	97	170	51	33	32	73	41	4	86	50	5	3	5	9	5	17	8	14
PhA	138	257	208	358	134	35	134	125	149	21	754	133	18	15	25	41	26	73	32	66
AnT	5	6	1	31	4	3	43	11	9	2	48	5	1	1	2	3	1	2	2	3
FluA	39	360	75	115	63	16	62	38	33	24	227	37	17	17	50	30	62	29	19	44
Pyr	46	90	41	60	67	6	26	56	58	43	109	38	36	36	75	69	107	37	31	60
BaA	1	9	1	5	2	2	10	2	1	3	2	1	2	1	7	1	2	1	1	4
Chr	11	334	16	46	20	5	32	8	7	10	69	11	6	5	42	9	21	9	5	15
BbF	5	8	1	2	25	5	30	2	2	9	7	1	6	3	34	2	7	2	3	7
BkF	5	10	5	8	7	2	11	4	6	7	5	3	5	1	20	9	9	3	1	9
BaP	6	10	4	7	22	6	9	10	13	28	4	2	94	4	12	19	15	1	2	19
InP	1	3	1	5	3	5	13	16	2	8	1	1	6	2	31	1	3	1	1	5
DbA	14	2	4	45	22	10	4	6	2	2	2	2	10	1	8	2	1	1	1	2
BghiP	6	4	11	96	38	11	15	22	5	14	6	4	19	3	30	12	4	3	2	7
ΣCPAH	32	43	16	71	81	30	77	39	27	57	22	10	123	12	111	34	37	9	10	46
ΣPAH	722	1,649	2,243	2,008	2,011	953	1,989	894	554	254	1,557	517	322	161	492	1,845	406	296	184	327
Moisture (%)	83.0	79.0	79.1	76.8	-	-	77.2	81.3	81.4	75.7	70.6	81.1	75.6	79.5	78	75	71.0	83.7	80.1	79.0
Lipid (%)	6.8	4.1	15.2	46.9	19.3	48.3	12.2	31.8	14.0	2.2	14.7	18.1	1.2	5.0	3	13	1.5	14.6	5.8	0.4

was also the highest among seafood samples. This seems to be relation to habitat surrounding. Namely, the flatfish, the benthic-dwelling species, is exposed to PAHs from contact with contaminated sediments by high-molecular-weight PAHs including benzo[a]pyrene. Therefore, heavier molecular weight PAHs such as benzo[a]pyrene in flatfish is thought to be a high contents.

The PAH levels in fishes measured in this study were compared with those in other countries (Table 3). There is little information on the level of PAHs in fishes in the world. Although the number of PAHs, fish species and sample types (muscle or liver) analyzed in other studies may differ, the sixteen PAHs have found in most samples regardless of fish species and sample types.

The sixteen PAH residues in 16 fish muscles excluding common squid, Korean shrimp, topshell, and blue crab varied from 0.2 to 2.2 ng/g wet weight with a mean value of 1.03 ng/g wet weight. These experimental results were similar or slightly low values compared to PAH levels in lake trout sampled from Great Lakes, ranging from 1.5 to 4.0 ng/g wet weight (Zabik et al., 1996). Sole et al. (2001) reported that 14 PAHs levels in deep-sea fish gadiform (*Mora moro*) from Mediterranean Sea varied from 0.2 to 0.6 ng/g wet weight in muscle and varied from 6.9 to 16 ng/g wet weight in liver. These residues in muscle were slightly lower than those in this study.

However, the PAH levels in yellowtail flounder (*Pleuronectes ferruginea*) tissues from Grand Bank were in the range of 0.5~34 ng/g wet weight with a mean value of 17.6 ng/g wet weight (Hellou and Warren, 1995). These values revealed an order of about 10~20 fold greater than those reported here. Also, other reports (Baumard et al., 1998; Valette-Silver et al., 1999; Pointet and Milliet, 2000; Sericano et al., 2001) in Table 3 seemed to be relatively greater than in this study despite the difference of sample type.

There was no correlation ($r=0.17$, $n=20$) between PAH residues and lipid contents in seafood samples. This may reflect relatively rapid changes in lipid content within individual fish as compared to the slow uptake and depuration of persistent organic contaminants including PAHs within fish species (Manchester-Neesvig et al., 2001).

Factor analysis of PAH profiles in marine fishes, sediments, bivalves and various marine organisms

For the investigation on similarities, differences, and relationships of the variations in PAH profiles

Table 3. Comparison of PAH residues in fish species of other countries in the world

Locations	Species	Type of sample	n[a]	Mean	Range	References
Korean coast	17 fish species	Muscle[b]	16	1.03	0.2~2.2	This study
Great Lakes, USA	Lake trout	Muscle[b]	14	3.25	1.5~4.0	Zabik et al., 1996
Mediterranean Sea	Gadiform	Muscle[b] Liver[b]	14	0.4 12.3	0.2~0.6 6.9~16	Solé et al., 2001
Mediterranean Sea	Sea comber, Red mullet	Liver[c]	14	41.3	14.7~139	Baumard et al., 1998
Nova Scotia, Canada	Yellowtail flounder, American plaice	Muscle[b] Liver[b]	16	17.6 10.2	0.5~3.4 1.5~2.3	Hellou and Warren, 1995
Kara Sea, Russia	Sturgeon	Liver[c]	18	590	420~760	Sericano et al., 2001
La Camargue, France	Goldfish, Eel, Catfish	Liver[b]	10	251	93~479	Pointet and Milliet, 2000
Western Beaufort Sea	Thule	Muscle[c]	18	67	24~161	Valette-Silver et al., 1999

[a]n: the number of analyzed PAHs.
[b]in ng/g wet weight.
[c]in ng/g dry weight.

in different sample matrices, factor analysis has been used (Kennicutt et al., 1992; Conde et al., 1996; Pena Mendez et al., 1996; Pettersen et al., 1997). The PAH profiles in fishes according to exposure pathway were compared using factor analysis corresponding to the PAH distributions in marine sediments, bivalves, crab, shrimp, and topshell samples. PAH data observed in marine sediments and bivalves (oysters and mussels) sampled from Korean coastal areas were used in this investigation (Moon et al., 2001b). The relationship plots between 16 variables (the individual PAH of 16 species) and the fish, sediment, bivalve, and various marine organism samples on the factorial plane are illustrated in Fig. 1.

On the variable map ((A) in Fig. 1), the individual PAH compound was used as variables in the factor analysis. The two loading factors accounted for 43.4% and 25.4% of the total variance of the original data, respectively. It could be noted that each factor is significantly correlated with a different group of compounds. The higher-molecular-weight aromatic compounds were correlated with loading factor 1, whereas the lower-molecular-weight aromatic compounds were correlated with loading factor 2. Circles in the variable map could be distinguished by three groups of PAHs according to the numbers of aromatic rings. The first group represented the lower-molecular-weight two and three ring aromatics, the most water soluable PAHs such as naphthalene, acenaphthylene, acenaphthene, fluorene, phenanthrene and anthracene except dibenzo [a,h]ahthracene. The second group comprised the four ring aromatic PAHs such as fluoranthene, pyrene, chrysene except benzo[g,h,i]perylene. Finally the third group consisted of the higher-molecular-weight five and six ring aromatic PAHs such as benzo[b]fluoranthene, benzo[k]fluoranthene, benzo [a]pyrene and indeno[1,2,3-c,d]pyrene except benzo [a]anthracene.

On the sample map ((B) in Fig. 1), various marine environmental samples were used as variables in the factor analysis. The two loading factors accounted for 48.5% and 27.7% of total variance in the data set, respectively. Four groups among the samples (defined by four circles) were to be distinguished and were related to PAH distribution in the sample type. The first group was integrated by variables of fish samples. The second cluster was integrated by variables of shrimp, crab and topshell species. The third one comprised variables of bivalve samples and finally a fourth one integrated by variables of marine sediment samples. Most of the PAHs within the same sample types were located close together near the circumference. This result suggested that the PAH profiles in each sample type for various marine environmental samples are similar and that most of the PAHs within the same samples behave identically in marine environment.

Representative profiles of PAHs in each marine environmental sample are illustrated in Fig. 2. As shown in Fig. 2, the contributions of ring aromatic

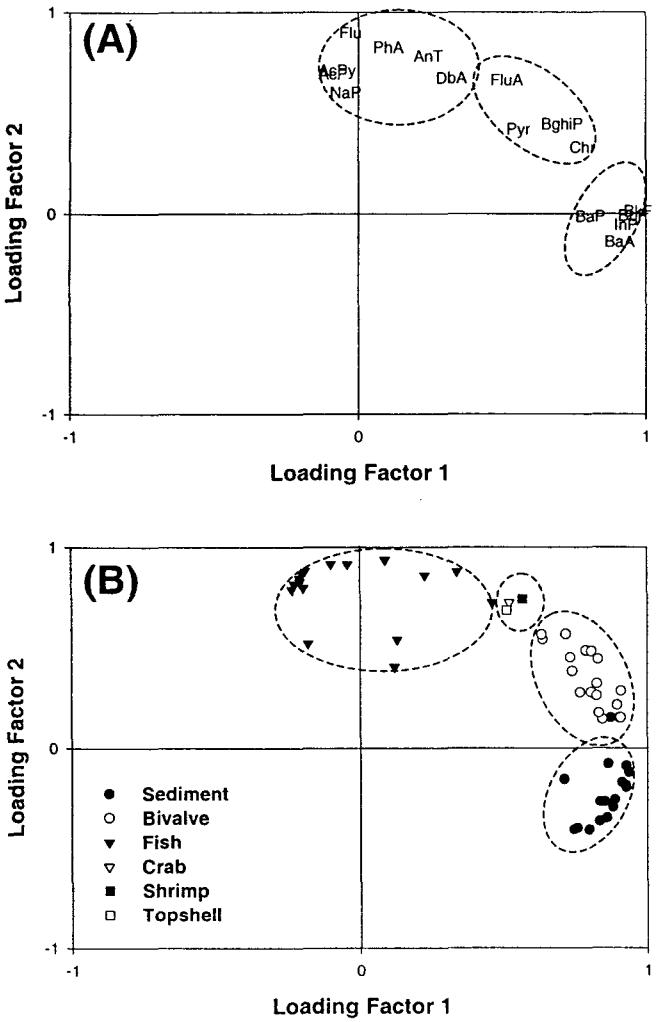

Fig. 1. Factor analysis of the sixteen PAH distribution for marine sediment, bivalve, fish, crab, shrimp and topshell collected from Korean coastal areas. (A) Variable map for relative sample matrix PAH concentrations as variables. (B) Sample map for the 17 sediments (●), 18 bivalves (○), 17 fish (▼), crab (▽), shrimp (■) and topshell (□).

groups to the sum of total PAHs in sediments were predominant in heavier molecular weight four and five ring aromatics like fluoranthene, pyrene, benzo [a]anthracene, chrysene, benzo[b]fluoranthene, benzo [k]fluoranthene and benzo[a]pyrene. This is due to their higher persistence and adsorption into sediment matrix based on more lipophilic and hydrophobic character (Witt, 1995; Benlahcen et al., 1998). In the bivalve samples, the predominant contributors were four ring aromatics such as fluoranthene, pyrene, benzo[a]anthracene and chrysene.

Contrasting with sediment and bivalve samples, the predominant PAHs in fish samples were lower -molecular-weight compounds such as two and three ring aromatics-naphthalene, acenaphthene, fluorene and phenanthrene-in accounting for more than about 80% of the total PAHs. Hydrophobic contaminants such as PAHs tend to rapidly adsorb on particles (Neff, 1979) and decreases as the octanol-water partition coefficient (K_{ow}) increases. PAH solubility decreases with increasing molecular weight (Porte and Albaigs, 1993; Djomo et al., 1996). Therefore, the lower-molecular-weight PAHs are preferentially dissolved, while the heavier molecular weight compounds are preferentially absorbed onto or associated with particles. Thus, the uptake of these contaminants is governed by its bioavailability, and fish is often enriched in the lower-molecular-weight PAHs relative to the sediment (Porte and Albaigs, 1993; Djomo et al., 1996; Baumard et al., 1998). In addition, the variations of the PAH profiles between fish and bivalve samples could be related to metabolic differences. Fish have a greater metabolic capacity relative to bivalves and biotransformation of the heavier molecular weight PAHs with a greater efficiency (Livingstone et al., 1992; Djomo et al., 1996). Hence, the PAH accumulation in fish tissue is mainly governed by the lower-molecular-weight PAHs.

The predominant PAHs in tissues of shrimp, crab and topshell were three- and four-ring aromatic compounds like acenaphthene, phenanthrene, fluoranthene and pyrene although there was a slight difference between species. The differences of PAH profiles in these marine organisms relative to fish could result from exposure pathway, different diets and biotranformation capacities of the organisms in addition to a different bioavailability of the compounds (Broman et al., 1990; Leonards et al., 1997). Namely, these marine species were accumulated by the bioconcentration of contaminants adsorbed in the sediment particle and dissolved in the water and by the biomagnification of preys they feed on. Therefore, the PAH profiles in shrimp, crab and topshell tissues were characterized by intermediate between those observed in the sediments and in the fish species.

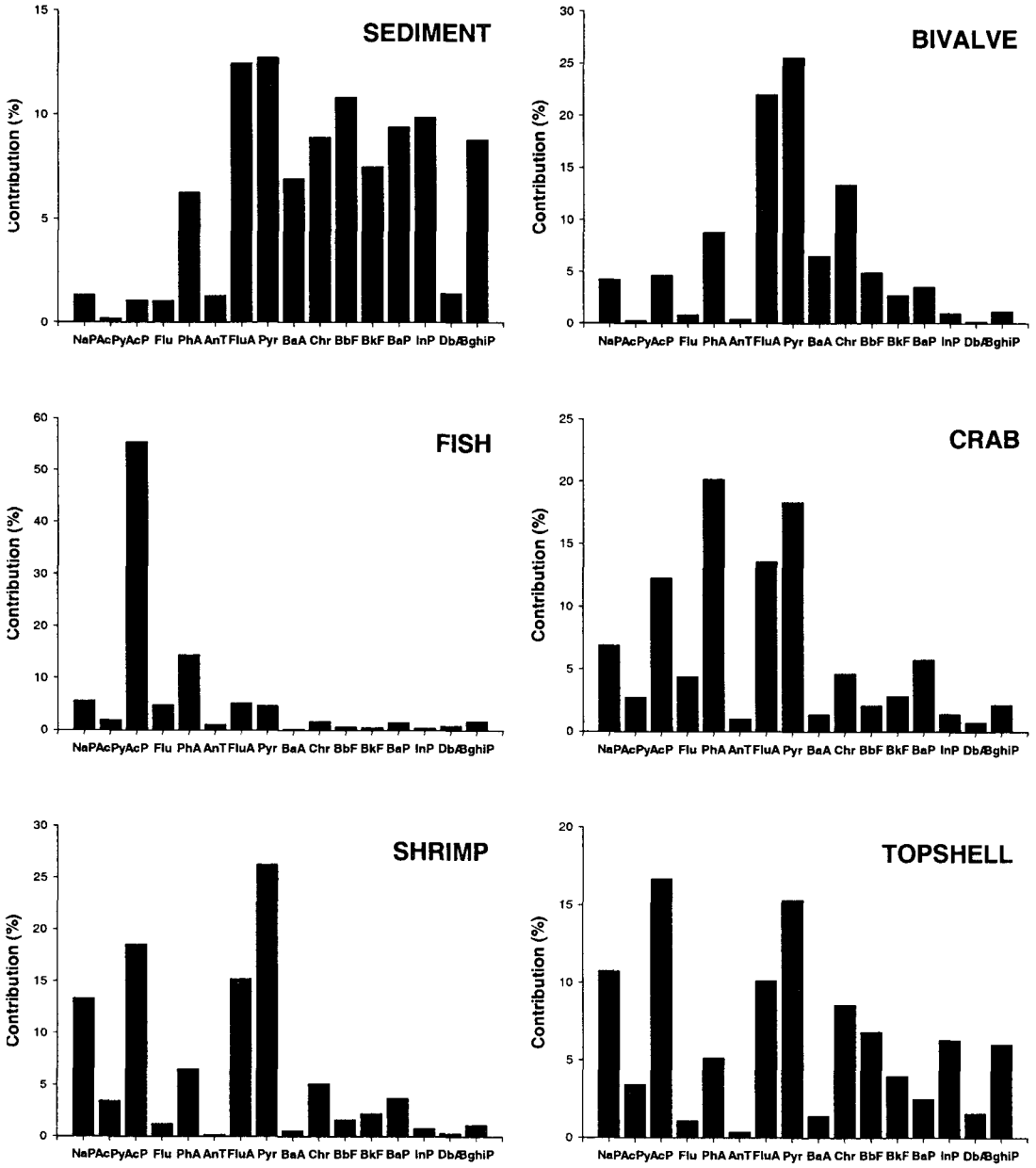

Fig. 2. Representative profiles of sixteen PAHs for marine sediments, bivalves, fishes, crab, shrimp and topshell.

Conclusions

Seafood was sampled at the local fisheries markets from Korean coastal areas to investigate residues and profile of polycyclic aromatic hydrocarbons (PAHs) accumulation. The levels of sixteen PAHs in seafood from Korean coast were 161 to 2,243 pg/g wet weight. The highest concentration was found at saury (*Coloabis saira*) and the lowest level was found at jacopever (*Sebastes schlegeli*). The residues of PAHs in fishes from Korean coasts were slightly low or relatively moderate to other countries. The predominant contributors in fish samples were lower-molecular-weight two and three ring aromatic PAHs, while filter-feeding organism like shrimp, crab and topshell were dominated by three- and four-ring aromatics. The PAH profiles in fishes according to exposure pathway were compared with marine sediments, bivalves (oysters and mussels) and benthic-feeding organisms using factor analysis. The PAH profiles in each sample species or mat-rices samples showed similar distribu-

tions. This result suggests that most of the PAHs within the same sample types behave identically in marine environ-ment.

Acknowledgements

The authors wish to thank Dr. H.G. Jin, Dr. J.I. Park and Mr. K.A. Jeon in East Sea Fisheries Research Institute, Mr. K.Y. Kim, Mr. J.S. Lee, Mr, J.N. Kwon, Mr. J.I. Choi and Mr. S.H. Cho in South Sea Fisheries Research Institute and Dr. K.H. An, Dr. G.S. Park and Mr. O.I. Choi in West Sea Fisheries Research Institute for assistance with sampling seafood.

References

Adamo, R.D., S. Pelosi, P. Trotta and G. Sansone. 1997. Bioaccumulation and biomagnification of polycyclic aromatic hydrocarbons in aquatic organisms. Mar. Chem., 56, 45~49.

Baumard, P., H. Budzinski, P. Garrigues, J.C. Sorbe, T. Bur-geot and J. Bellocq. 1998. Concentrations of PAHs (Poly-cyclic aromatic hydrocarbons) in various marine organi-sms in relation to those in sediments and to trophic level. Mar. Pollut. Bull., 36, 951~960.

Baumard, P., H. Budzinski, P. Garrigues, H. Dizer and P.D. Hansen. 1999. Polycyclic aromatic hydrocarbons in recent sediments and mussels (*Mytilus edulis*) from the Western Baltic Sea: occurrence, bioavailability, and seasonal variations. Mar. Environ. Res., 47, 17~47.

Benlahcen, K.T., C.H. Budzinski, J. Bellocq and P.H. Garrigues. 1998. Distribution and sources of polycylic aromatic hydrocarbons in some Mediterranean coastal sediments. Mar. Pollut. Bull., 34, 298~305.

Broman, D., C. Nf, I. Lundbergh and Y. Zebuhr. 1990. An in situ study of the distribution, biotransformation and flux of polycyclic aromatic hydrocarbons (PAHs) in an aquatic food chain (seston-*Mytilus edulis* L.-*Somateria mollissima* L.) from the Baltic: An ecotoxicological perspective. Environ. Toxicol. Chem., 9, 429~442.

Burkhard, L.P. and M.T. Lukasewycz. 2000. Some bioaccumulation factors and biota-sediment accumulation factors for polycyclic aromatic hydrocarbons in lake trout. Environ. Toxicol. Chem., 19, 1427~1429.

Conde, J., E. Pena and F.G. Montelongo. 1996. Sources of tar balls and oil slicks on the coasts of the Canary Islands. Inter. J. Environ. Anal. Chem., 62, 77~84.

Djomo, J.E., P. Garrigues and J.F. Narbonne. 1996. Uptake and depuration of polycyclic aromatic hydrocarbons from sediment by the zebrafish (*Bracydanio rerio*-). Environ. Toxicol. Chem., 15, 1177~1181.

Escartn, E. and C. Porte. 1999. Assessment of PAH pollution in coastal areas from the NW mediterranean through the analysis of fish bile. Mar. Pollut. Bull., 38, 1200~1206.

Fairey, R., K. Taberski, S. Lamerdin, E. Johnson, R.P. Clark, J.W. Downing, J. Newman and M. Petreas. 1997. Organochlorines and other environmental contaminants in muscle tissue of Sportfish collected from San Francisco Bay. Mar. Pollut. Bull., 34, 1058~1071.

Hellou, J. and V. Zitko, J. Friel and T. Alkanani. 1996. Distribution of elements in tissues of yellowtail flounder *Pleuronectes ferruginea*. Sci. Tot. Environ., 181, 137~146.

Hellou, J. and W.G. Warren. 1996. Polycyclic aromatic hydrocarbons in tissues of flatfish- insight on environmental exposure. Mar. Environ. Res., 43, 11~24.

Hendriks, A.J., H. Pieters and J. De Boer. 1998. Accumulation of metals, polycyclic (halogenated) aromatic hydrocarbons, and biocides in zebra mussel and eel from the Rhine and Meuse rivers. Environ. Toxicol. Chem., 17, 1885~1898.

Hites, R.A., R.E. LaFlamme and J.W. Farrington. 1977. Sedimentary polycyclic aromatic hydrocarbons- the historical report. Science, 198, 829~831.

Kennicutt, M.C., T.J. McDonald, G.J. Denoux and S.J. McDonald. 1992. Hydrocarbon contamination on the Antarctic Peninsula. II. Arthur harbor inter- and sub- tidal limpets (*Nacella concinna*). Mar. Pollut. Bull., 24, 506~511.

Kim, G.B., K.A. Maruya, R.F. Lee, J.H. Lee, C.H. Koh and S. Tanabe. 1999. Distribution and sources of polycyclic aromatic hydrocarbons in sediments from Kyeonggi Bay, Korea. Mar. Pollut. Bull., 38, 7~15.

Law, R.J. and J.L. Biscaya. 1994. Polycyclic aromatic hydrocarbons (PAHs) problems and progress in sampling, analysis and interpretation. Mar. Pollut. Bull., 29, 235~241.

Lee, R.F. and D.S. Page. 1997. Petroleum hydrocarbons and their effects in subtidal regions after major oil spills. Mar. Pollut. Bull., 34, 928~940.

Lee, K.S., I. Noh, C.S. Lim and S.D. Chu. 1998. The high performance liquid chromatography (HPLC) analysis of polycyclic aromatic hydrocarbons (PAHs) in oysters from the intertidal and subtidal zones of Chinhae Bay, Korea. J. Kor. Soc. Environ. Sci., 2, 57~68.

Lehr, R.E. and D.M. Jerina. 1977. Metabolic activations of polycyclic aromatic hydrocarbons. Arch. Toxicol., 39, 1~6.

Leonards, P.E.G., Y. Zierikzee, U.A. Brinkman, W.P. Cofino, N.M. Van Straalen and B. Van Hattum. 1997. The selective diaretary accumulation of planar polychlorinated biphenyls in the otter (*Lutra lutra*). Environ. Toxicol. Chem., 16, 1807~1815.

Livingstone, D.R. 1992. Persistent pollutants in marine invertbrates. In Persistent Pollutants in Marine Ecosystems, C. H. Walker and D.R. Livingstone, ed. Pergamon Press, Oxford, pp. 3~34.

Manchester-Neesvig, J.B., K. Valters and W.C. Sonzogni. 2001. Comparison of polybrominated diphenyl ethers (PBDEs) and polychlorinated biphenyls (PCBs) in Lake Michigan salmonids. Environ. Sci. Technol., 35, 1072~1077.

Mix, M.M. 1986. Cancerous diseases in aquatic animals and their association with environmental pollutants: a critical literature review. Mar. Environ. Res., 20, 141.

Moon, H.B., H.G. Choi, S.S. Kim, P.J. Kim, P.Y. Lee and G. Ok. 2001a. Polycyclic aromatic hydrocarbons in sediments from the southeastern coastal areas of Korea. J. Kor. Soc. Oceanogr., 36, 27~33.

Moon, H.B., H.G. Choi, S.S. Kim, S.R. Jeong, P.Y. Lee and G. Ok. 2001b. Monitoring of polycyclic aromatic hydrocarbons in sediments and organisms from Korean coast. J. Fish. Sci. Tech., 4, 219~228.

Neff, J.M. 1979. Polycyclic aromatic hydrocarbons in the aquatic environment, sources, fates and biological effects. Applied Science. London. UK., 262pp.

Pena Mendez, E.M., M.S. Astoga-Espana and F.J. Garcia-Montelongo. 2001. Chemical fingerprinting applied to the evaluation of marine oil pollution in the coasts of Canary Islands (Sapin). Envrion. Pollut., 111, 177~187.

Pettersen, H., C. Naf and D. Broman. 1997. Impact of PAH outlets from an oil refinery in the receiving water area - sediment trap fluxes and multivariate statistical analysis. Mar. Pollut. Bull., 34, 85~95.

Pointet, K. and A. Milliet, 2000. PAHs analysis of fish whole gall bladders and livers from the natural reserve of Camargue by GC/MS. Chemosphere, 40, 293~299.

Porte, C. and J. Albaigs. 1993. Bioaccumulation patterns of hydrocarbons and polychlorinated biphenyls in bivalves, crustaceans and fishes. J. Arch. Environ. Contam. Toxicol., 26, 273~281.

Prahl, F.G. and R. Carpenter. 1983. Polycyclic aromatic hydrocarbon (PAH) - phase associations in Washington coastal sediment. Geochem. Cosmo. Acta., 47, 1013~1023.

Sericano, J.L., J.M. Brooks, M.A. Champ, M.C. Kennicutt and V.V. Makeyev. 2001. Trace contaminant concentrations in the Kara Sea and its adjacent rivers, Russia. Mar. Pollut. Bull., 42, 1017~1030.

Soclo, H.H., P.H. Garrigues and M. Ewald. 2000. Origin of polycyclic aromatic hydrocarbons (PAHs) in coastal marine sediments: Case studies in Cotonou (Benin) and Aquitaine (France) areas. Mar. Pollut. Bull., 40, 387~396.

Solé, M., C. Porte and J. Albaigés. 2001. Hydrocarbons, PCBs and DDT in the NW Mediterranean deep-sea fish Mora moro. Deep-Sea Res., 48, 495~513.

Valette-Silver, N., M.J. Hameedi, D.W. Efurd and A. Robertson. 1999. Status of the contamination in sediments and biota from the western Beaufort Sea (Alaska). Mar. Pollut. Bull., 38, 702~722.

Wakeham, S.G., C. Schaffner and W. Giger. 1980. Polycyclic aromatic hydrocarbons in recent lake sediments: I. Compounds having anthropogenic origins. Geochem. Cosmo. Acta., 44, 403~413.

White, K.L. 1986. An overview of immunotoxicology and carcinogenic polycyclic aromatic hydrocarbons. Environ. Carcino. Rev., 2, 163~202.

Witt, G. 1995. Polycyclic aromatic hydrocarbons in water and sediment of the Baltic Sea. Mar. Pollut. Bull., 31, 237~248.

Yan, L.-S. 1985. Study of the carcinogenic mechanism for polycyclic aromatic hydrocarbons - extent bay region theory and its quantitative model. Carcinogenesis, 6, 1~6.

Zabik, M.E., A. Booren, M.J. Zabik, R. Welch and H. Humphrey. 1996. Pesticide residues, PCBs and PAHs in baked, charbroiled, salt boiled and smoked Great Lakes lake trout. Food Chem., 55, 231~239.