

Dietary Intake and Potential Health Risk of Polycyclic Aromatic Hydrocarbons (PAHs) via Various Marine Organisms in Korea

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Sixteen polycyclic aromatic hydrocarbons (PAHs) were analyzed in seventy marine organisms (40 species) from the Korean coast. PAHs were present in all the organisms. The level of total PAHs in the organisms varied from 0.45 to 224 ng/g dry weight and the carcinogenic PAHs varied from 0.05 to 49.8 ng/g dry weight. The PAHs residues according to the marine organisms showed a highest content in bivalve species, and followed by crustaceans, cephalopods, fish and gastropods. Human dietary intake of total PAHs through marine organism in Korea was estimated to be 4.12 ng/kg body weight/day and 0.67 ng/kg body weight/day for carcinogenic PAHs. The relative contributions of individual species to the total dietary intake of PAHs were in the order of bivalves (53.4%), fish (21.9%), crustaceans (15.3%), cephalopods (8.8%) and gastropods (0.6%). Daily dietary intake of PAH-TEQ expressed as a TEQ value was estimated to be 0.13 pg TEQ/kg body weight/day, which did not exceed a tolerable daily intake (TDI) proposed by the KFDA and the WHO as well as the UK toxicity committee. Lifetime cancer risk due to ingestion of marine species by the Korean adult was evaluated using the equation estimating exposure of food ingestion. Although approximately 23% of cumulative frequency of the sampled marine species exceeded the cancer risk guideline, lifetime cancer risk associated with marine organism consumption was negligible. Results indicate that dietary intake of PAHs through the consumption of the Korean marine organisms seems to be safe for human ingestion with negligible cancer risk.

Key words: PAHs, Marine organisms, Dietary intake, Lifetime cancer risk, Human health

Introduction

Polycyclic aromatic hydrocarbons (PAHs) are toxic, carcinogenic and mutagenic to all organisms including humans (Yan, 1985; Mix, 1986), and they are widespread pollutants in marine ecosystems and occur mainly as a result of anthropogenic activities (Adamo et al., 1997; Hendriks et al., 1998). The main sources of these contaminants are byproducts of incomplete fuel combustion (Hites et al., 1977), domestic and industrial wastewaters (Wakeham et al., 1980), and spillage of crude oil and its refined products (Lee and Page, 1997; Pettersen et al., 1997). Toxic organic contaminants are primarily transported into the ocean via two pathways, atmospheric deposition and direct and/or river inputs.

Marine organisms may be exposed to PAHs through

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 hydrophobic character and low biodegradability of PAHs cause bioaccumulation in marine organisms beyond their concentrations from the surrounding environment (Van der Oost et al., 1988; Oliver and Niimi, 1988).

PAHs are known to occur in aquatic environments and accumulate in aquatic organisms, so that consumption through marine organisms may be a significant human dietary source of these chemicals eventually posing a potentially real health risk (Fairey et al., 1997; Sericano et al., 2001). Seafood is the primary source of protein for people in the world and an important part of diet in Korean population. People can be exposed to toxic chemicals that accumulate in contaminated seafood which they consumed

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(Han et al., 1994; Svensson et al., 1995). The exposure of humans to toxic contaminants occurs mainly (>95%) through the contaminated food. The other exposure pathways, however, contribute bioaccumulation to a negligible extent (Connet and Webster, 1987; Abad et al., 2002). Hence, determination of the concentration of these compounds in various marine species are very important for evaluation of toxic dietary exposure, particularly due to the increase of seafood consumers. The purpose of the present study is to estimate the dietary intake of PAH via various popular marine species throughout the Korean coast and to evaluate potential human health risks by their consumption.

Materials and Methods

Sample collection

Seventy marine organism samples (40 species) were purchased at local fish markets from various Korean coastal areas from January 2002 to October 2003. These organisms are commonly consumed species and are commercially important in Korea. The selected marine species can be classified into 5 groups including fish (38 samples, 24 species), crustaceans (9 samples, 5 species), bivalves (13 samples, 6 species), gastropods (6 samples, 3 species) and cephalopods (4 samples, 2 species). Detailed biological information on sampled organisms in this study was reported in Moon et al. (2004).

Sample preparation and analysis

Marine organism samples were stored in a cooler box with ice or dry ice and immediately transported to the laboratory. The samples were homogenized with an ultra-disperser and then freeze-dried. Internal standards of 7 species (ES 2044, Cambridge Isotope Laboratories, Inc.) were spiked into freeze-dried samples (approximately 3 g) and were then digested in 150 mL of 1 N KOH ethanolic solution for 2 hours by mechanical shaking. The digest was liquid-liquid extracted twice using 150 mL of *n*-hexane (Ultra residue analysis, J.T. Baker) after the addition of water and 50 g of anhydrous Na₂SO₄. The extracts were reduced to a small volume in a rotary evaporator and then adjusted to a volume of 10 mL.

Extracts of marine organisms were cleaned using activated silica gel (Art no. 7734, 70-230 mesh, Merck) columns successively eluted with *n*-hexane and 15% methylene dichloride (Dioxin analysis, Wako) in *n*-hexane. The second fraction was con-

centrated to less than 1 mL, and left at a room temperature for one day to evaporate to 100-200 μ L. The residues were dissolved in 100 μ L of *n*-nonane (Pesticide residue analysis, Fluka) and analyzed for PAHs with GC/MSD (Agilent 5973N, USA). Further details of the experimental procedure and instrumental analysis of PAHs were presented in Moon et al. (2001, 2002).

The 16 non-alkylated PAH compounds recommended as the toxic priority pollutants by the United States Environmental Protection Agency (US EPA) were analyzed for each organism sample. These were as follows: 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).

QA/QC

All the spiked internal standards were detected with no interfering peak. The average recoveries showed 76 \pm 7% for two-, 75 \pm 6% for three-, 82 \pm 7% for four-, 87 \pm 4% for five-, and 92 \pm 6% for six-ring aromatic groups.

The certified mussel (*Mytilus edulis*) homogenate (1974a, NIST, USA) was analyzed as a Standard Reference Material (SRM) in this study, in order to assess the accuracy of the results of experimental procedure and instrumental analysis. Average recoveries for two-, three-, four-, five-, and six-ring aromatic groups were 67%, 78%, 84%, 91% and 92%, respectively. Procedural blanks were processed in the same manner as real samples, and they were all below 10% of analytes abundance. Blanks were run before and after the injection of standard solutions to check for any carryover.

Calculation of dietary intake

The average body weight of the Korean adult was set as 60 kg. The data on average daily dietary intake were obtained from the Ministry of Health and Welfare (MOHW, 2001). Total average daily ingestion through foods of Korea was 1,315 g. Seafood was 64.1 g or 5% of total food ingestion. This value included both natural and processed seafoods. The ingestion values were calculated as the sum of all individual sampled natural species. PAHs data for daily dietary intake were expressed as a wet weight

basis recalculated by moisture content (%) of each organism in this study. Daily dietary intake of PAHs through consumption of various marine species was calculated by multiplying the concentration of contaminants in organism samples (ng/g wet weight basis) by the samples available amounts of individual marine species (average daily intake in g/day).

Results and Discussion

Concentration of PAHs in various marine species

The concentrations of PAHs in various marine species from coastal regions of the Korean coast were summarized in Table 1. The moisture and lipid contents (dry weight basis) in the organisms were in the ranges of 46-84% and 1.1-48%, respectively. Fish species particularly showed the widest range of lipid content with 1.1-48% (Table 1). All the 16 PAHs were detected in all organism samples collected. The levels of Σ PAH (the sum of each PAH) in all kinds of the organisms were in the range of 0.45-224 ng/g dry weight and total concentrations of potentially carcinogenic PAHs (Σ CPAH; the sum of BaA, BbF, BkF, BaP, InP and DbA) (IARC, 1987) were in the range of 0.05-49.8 ng/g dry weight.

Fish species had the widest bioaccumulation range of PAHs in comparison to other species. Concentrations of Σ PAH in fish species varied from 0.45 to 56.0 ng/g dry weight and Σ CPAH varied from 0.05 to 5.56 ng/g dry weight. The highest contents were detected in rockfish (*Sebastes olivaceus*) from the Incheon coast. Sailfin sandfish (*Arctoscopus japonicus*) from the Jumunjin coast and roundnose flounder (*Eopsetta grigorjewi*) from the Pohang coast also showed relatively high contents of PAHs. Residues of Σ PAH in crustacean species were in the range of 1.40-71.8 ng/g dry weight, and Σ CPAH were in the range of 0.13-49.8 ng/g dry weight. Red snow crab (*Chionoecetes japonicus*) and the hair crab (*Erimacrus isenbecki*) showed the higher residues

of PAHs than other species. The levels of Σ PAH in bivalve species ranged from 13.9 to 224 ng/g dry weight, and Σ CPAH ranged from 2.42 to 39.2 ng/g dry weight. Mussel (*Mytilus coruscus*, 224 ng/g dry weight) from the Gwangyang coast and the Pacific oyster (*Crassostrea gigas*, 137 ng/g dry weight) from the Incheon coast contained the highest levels of PAHs. For gastropod species the levels of Σ PAH varied from 1.31 to 18.2 ng/g dry weight and Σ CPAH varied levels from 0.19 to 4.28 ng/g dry weight. Concentrations of Σ PAH and Σ CPAH in cephalopod species showed the low residues.

Bivalve species showed the highest contents of PAHs in this study (Fig. 1). Marine bivalves can bioaccumulate xenobiotics adsorbed on fine particles, which contain the highest concentrations of PAHs. Hence, bivalves such as the oyster and the mussel have been used bio-monitoring species for PAHs contamination in the coastal environments of Korea and other countries by the previous works (Moon et al., 2001; Goldberg et al., 1978; Philips, 1978). Fish species with high trophic level showed the lowest concentrations in comparison to other species. This is due to the high metabolism of fish relative to the other organism species. This indicates that PAHs migrated into fish can be eliminated by biotransformation and/or biodegradation with greater efficiency (Broman et al., 1990; Leonards et al., 1997). Baumard et al. (1998) reported that marine organisms of high trophic levels can partly biotransform some contaminants such as PAHs.

Dietary intake of PAHs via various marine species

The daily dietary intake for Σ CPAH, Σ PAH and PAH-TEQ according to marine species were calculated as shown in Table 2. The dietary intake of Σ CPAH for various marine species was 0.67 ng/kg body weight/day and that for Σ PAH was 4.12 ng/kg body weight/day. Daily dietary intake of Σ CPAH and Σ PAH via fish consumption was 0.12 and 1.21

Table 1. Concentration (ng/g dry weight) of PAHs in various marine organisms from the coastal areas of Korea

	Moisture (%)	Lipid (%) ¹	Σ PAH	Σ CPAH
Fish ($n^2=38$)	62 - 84	1.1 - 48	0.45 - 56.0	0.05 - 5.56
Crustacean ($n=9$)	58 - 79	2.4 - 17	1.40 - 71.8	0.13 - 49.8
Bivalve ($n=13$)	46 - 84	2.0 - 10	13.9 - 224	2.42 - 39.2
Gastropod ($n=6$)	65 - 78	2.1 - 4.0	1.31 - 18.2	0.19 - 4.28
Cephalopod ($n=4$)	63 - 79	3.1 - 4.7	3.60 - 19.7	0.20 - 2.23

Lipid¹ in the dry weight; n^2 : sample numbers.

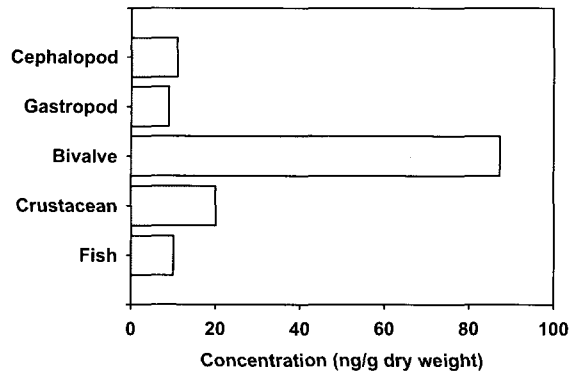


Fig. 1. Comparison of average concentrations of Σ PAH according to various marine species from the coastal areas of Korea.

ng/kg body weight/day, respectively. Crustacean's daily intake for Σ CPAH and Σ PAH were 0.11 and 0.32 ng/kg body weight/day, respectively. Bivalve species was characterized by the highest exposure to PAHs with 0.39 and 2.15 ng/kg body weight/day for Σ CPAH and Σ PAH, respectively. Gastropod and cephalopod species showed relatively low ingestion of PAHs.

The carcinogenic induction potency value proposed by Klimm et al. (1999) was used, to get a tolerable daily intake (TDI) expressed as total equivalents (TEQ) value through ingestion of seafood contaminated with PAHs. Toxic equivalent factor (TEF) values used for each compound were BaA (0.000027), BbF (0.00038), BkF (0.00029), BaP (0.0003), InP (0.000086), and DbA (0.000078). The dietary intake of PAH-TEQ expressed as a TEQ value was 0.13 pg TEQ/kg body weight/day (Table 2). Dietary intake of PAH-TEQ in fish was 0.029 pg TEQ/kg body weight/day. Crustacean's dietary intake for PAH-TEQ was 0.02 pg TEQ/kg body weight/day. In bivalve

species, it was 0.07 pg TEQ/kg body weight/day. Gastropod and cephalopod species showed relatively low dietary intake of PAHs in this study.

Recently, the WHO expert committee recommended a TDI of 1-4 pg TEQ/kg body weight/day (SCF, 2001). In addition, a TDI for human exposure in the Korean population was proposed as 4 pg TEQ/kg body weight/day by the Korea Food Drug and Administration (KFDA). Most recently, the UK committee on toxicity of chemicals in food, consumer products and the environment has recommended a TDI of 2 pg TEQ/kg body weight/day (CoT, 2001). In this study, the estimated dietary intake of PAHs (0.13 pg TEQ/kg body weight/day) by consumption of marine species did not exceed the TDIs proposed by the KFDA, the WHO, and the UK toxicity committee. Therefore, the dietary intake of PAHs by the consumption of Korean marine organisms was relatively safe for human health risk.

The relative contribution of individual species to the total dietary intake of PAHs (Fig. 2) was highest

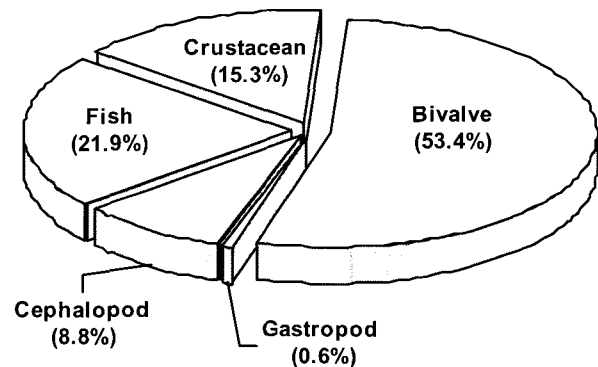


Fig. 2. Relative contribution to the estimated dietary intake of various marine species for TEQ values (pg TEQ/kg body weight/day) of PAHs.

Table 2. Estimated daily dietary intake for Σ CPAH, Σ PAH and PAH-TEQ via various marine species from coastal regions of Korea

	Average intake (g/day)	Daily dietary intake		
		Σ CPAH (ng/kg body weight/day)	Σ PAH (ng/kg body weight/day)	PAH-TEQ ¹ (pg TEQ/kg body weight/day)
Fish	30.5	0.12	0.21	0.029
Crustacean	3.3	0.11	0.319	0.020
Bivalve	4.4	0.39	2.15	0.070
Gastropod	0.5	0.004	0.021	0.001
Cephalopod	7.8	0.04	0.418	0.012
Total	46.5	0.67	4.12	0.13

¹TEQ values for PAHs were calculated as the carcinogenic induction potency value proposed by Klimm et al. (1999).

Table 3. Oral slope factors for carcinogenic PAHs in fish

Chemicals	CAS number	Oral slope factor (mg/kg/day) ⁻¹	Reference
Benzo[a]anthracene	56553	0.73	US EPA (2004)
Chrysene	218019	0.0073	US EPA (2004)
Benzo[b]fluoranthene	205992	0.73	US EPA (2004)
Benzo[k]fluoranthene	207089	0.073	US EPA (2004)
Benzo[a]pyrene	50328	7.3	US EPA (2004)
Indeno[1,2,3-c,d]pyrene	193395	0.73	US EPA (2004)
Dibenzo[<i>a,h</i>]anthracene	53703	7.3	US EPA (2004)

amongst bivalves (53.4%), followed by fish (21.9%), crustaceans (15.3%), cephalopods (8.8%) and gastropods (0.6%). Although fish had the highest ingestion amounts compared to the other marine species, the relative contribution to total dietary intake was relatively low residues of PAHs.

Lifetime cancer risk

Health risk to human from ingesting toxic contaminants is classified as carcinogenic and non-carcinogenic. In this study, the potential health risks of ingesting marine species contaminated with carcinogenic environmental pollutants were evaluated for the Korean adult population using the risk assessment guideline of the US EPA. The general equation for estimating exposure via ingestion of food items is as follows:

$$\text{Cancer Risk} = \frac{I \cdot ED \cdot EF \cdot CSF}{BW \cdot AT \cdot 365} \quad (\text{US EPA, 1989}),$$

where *I* is the daily dietary intake (mg/kg/day); *ED* is the exposure duration (adults=30 years); *EF* is the exposure frequency (350 days/year); *CSF* is the oral cancer slope factor (data for individual chemicals was obtained from IRIS reported by US EPA (2004)) (Table 3); *BW* is human body weight (60 kg); *AT* is the average time for carcinogens (365 days/year × number of exposure years, assuming 70 years). From this equation, a cancer risk value below 1×10^{-6} indicates that the level of exposure is not likely to cause any adverse effect in the Korean human population during their lifetime.

Lifetime cancer risk values from the dietary intake of each tested marine species were estimated for the Korean adult population using the foregoing equation (Table 4). The highest toxic effect was from bivalvia species with 4×10^{-7} . However, none of the marine organisms posed a lifetime cancer risk. The sum of cancer risk (9×10^{-7}) for all marine organisms did not exceed a cancer risk guideline value of 1×10^{-6} .

The cumulative frequency of the lifetime cancer

Table 4. Lifetime cancer risks from dietary intake of contaminated marine species in the present study

	Lifetime cancer risk
Fish	2×10^{-7}
Crustacean	1×10^{-7}
Bivalve	4×10^{-7}
Gastropod	8×10^{-9}
Cephalopod	7×10^{-8}
Sum	9×10^{-7}

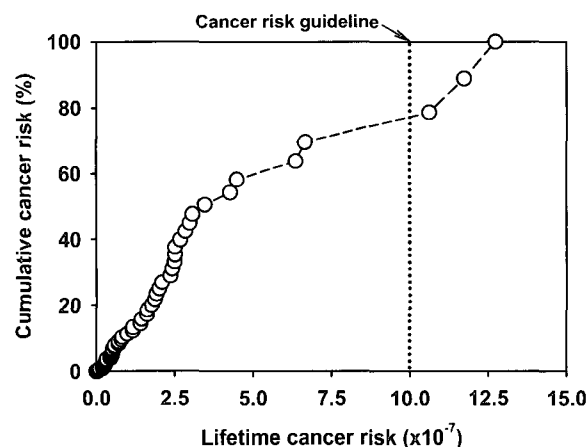


Fig. 3. Cumulative plot of the lifetime cancer risk for PAHs by consumption of marine organism from coastal regions of Korea.

risk via marine species consumption was presented in Fig. 3. Approximately 23% of marine species exceeded the cancer risk guideline. Some fish and crustaceans were highly contaminated by PAHs, and thus the values exceeded the cancer risk guideline. However, most marine organisms were below the cancer risk guideline with 4×10^{-7} . These results indicate that cancer risk is negligible through the consumption of the Korean marine organisms during a lifetime of the Korean population. However, more information should be obtained on potential health risks of various toxic chemicals on human at low

exposure levels, particularly in neurodevelopment. In addition, an adequate guideline for dietary intake of marine organisms should be established in Korea, to protect and manage the public health in Korea.

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