# Monitoring of Polycyclic Aromatic Hydrocarbons in Sediments and Organisms from Korean Coast

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Surface sediments (0~5 cm), mussels (*Mytilus coruscus* and *M. edulis*) and oysters (*Crassostrea gigas*) were sampled at 20 stations in Korean coast during a period of February to July 2000. Samples were analyzed for polycyclic aromatic hydrocarbons (PAHs) content using gas chromatography coupled to mass spectrometer detector (GC/MSD). The sediment $\Sigma$PAH concentration varied from 7 to 1,214 ng/g dry weight and the level of carcinogenic PAHs of six species ranged from 1 to 563 ng/g dry weight in surface sediments. In organisms, the levels of $\Sigma$PAH were in the range of 63~876 ng/g dry weight and the concentrations of carcinogenic PAHs of six species were in the range of 4~582 ng/g dry weight. The highest PAH concentrations in sediments and organisms were in samples from Station 5 in Pohang coast and Station 8 in Jinhae coast, respectively. $\Sigma$PAH concentrations in sediments and organisms collected from Korean coast were slightly low or comparable to those in other countries. The contribution of ring aromatic groups to sum of 16 PAHs in sediments and organisms showed a similar pattern for most stations. In sediments, the predominant contributions were four and five ring aromatics like fluoranthene, pyrene, benzo[a]anthracene, chrysene, benzo[b]fluoranthene, benzo[k]fluoranthene and benzo[a]pyrene. In the organisms, four ring aromatics like fluoranthene, pyrene, benzo[a]anthracene and chrysene were predominant PAH species. Several molecular indices such as phenanthrene/anthracene, fluoranthene/pyrene and LMW/HMW ratio were used to identify the origin of PAH contamination in sediments. The results indicate that PAHs of Korean coast were mainly of pyrolytic contaminants with slight petrogenic input. A high correlation within individual PAH compound in marine sediments was observed, while correlation in organisms showed a highly significant relationship for heavier molecular weight PAHs.

Key words: Sediment, Mussel, Oyster, Polycyclic aromatic hydrocarbons (PAHs), Contribution, Pyrolytic, Petrogenic

## Introduction

In recent years, there has been an increasing emphasis on the study of the environmental fate and effects of polycyclic aromatic hydrocarbons (PAHs). PAHs are well-known ubiquitous contaminants of the coastal marine environment. These compounds have mainly anthropogenic origins (Wakeham et al., 1980), including combustion process of fossil

fuels (Hites et al., 1977), domestic and industrial wastewater (Wakeham et al., 1980) and spillage of petroleum or petroleum products by ships (Lee and Page, 1997; Pettersen et al., 1997). Each source is characterized by a specific molecular pattern and it is, therefore, possible to identify which source generated these chemicals. PAHs derived from various sources transport to the marine environment via both waters and atmosphere. Most PAHs in water column tend to be adsorbed to particles and to be deposited to the underlying sediments (Law and

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Biscaya, 1994). Degradation of PAHs in sediment is generally slow, particularly for the higher-molecular-weight PAH (Readman et al., 1982). This class of compounds in marine sediments and organisms has been widely studied (Neff, 1979) because of its carcinogenic and mutagenic character (Lehr and Jerina, 1977; Yan, 1985; Mix, 1986; White, 1986).

Aquatic organism may bioaccumulate and bioconcentrate environmental contaminants to more than 1,000,000 times of the concentrations detected in the water column (Oliver and Niimi, 1988; Spies et al., 1988). International monitoring programs for marine pollutants are carried out using bioindicators such as shellfish, particularly mussels (*Mytilus edulis*) and oysters (*Crassostrea gigas*) (Goldberg et al., 1978; Phillips, 1978). For example, the Mussel Watch Program in the USA and the Reseau National d'Observation (RNO) in France have been developed using mussel and oysters to monitor spatial and temporal trends of contaminant concentration in coastal and estuarine regions (Farrington et al., 1983; Claisse et al., 1992; O'Connor, 1996). These bivalves are an effective trapping mechanism for many environmental pollutants (PCBs, *p,p'*-DDE, PAHs, metals, etc.) because of their wide distribution, abundance, sedentary behavior, and pronounced ability to accumulate organic compounds at proportions that correspond to concentrations in their surroundings. Moreover, analysis of bivalve tissue gives an indication of the bioavailable fraction of environmental contamination and of direct exposure to chemicals. In the present study, we planned to monitor PAHs pollution in marine sediments and organisms collected from several coastal areas in Korea.

## Materials and Methods

Surface sediments (0~5 cm) and organisms were sampled at 20 stations in Korean coastal areas during a period of February to July 2000 (Fig. 1). Sediments were collected either with a box-core sampler. Mussels (*Mytilus coruscus* and *M. edulis*) and oysters (*Crassostrea gigas*), which were located on piers, rocks or buoys in the water column, were scraped with a rake. The shells of the mussels and oysters were removed carefully. Both sediments and bivalve tissues were wrapped with an aluminum foil
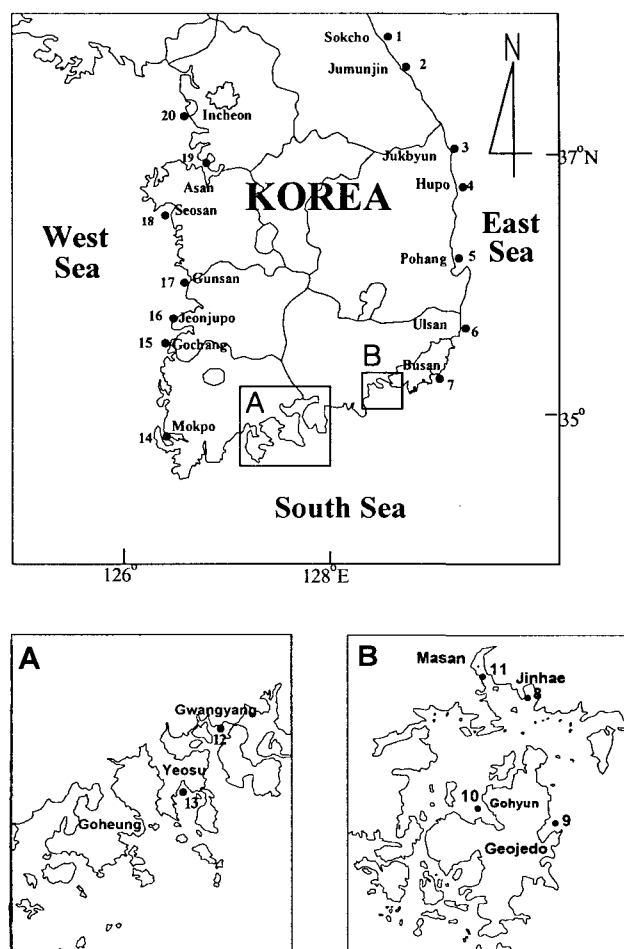


Fig. 1. Map showing sampling stations of marine sediments and organisms from Korean coast.

and frozen at −20℃.

The freeze-dried sediments were sieved at 2 mm and then extracted in a Soxhlet apparatus with 200 mL of toluene (Ultra residue analysis, J.T. Baker) for 20 hours after the spike of seven species of internal standard (ES 2044, Cambridge Isotope Laboratories, Inc.), then the volume was reduced to 1~2 mL in a rotary evaporator. The extracts were transferred to *n*-hexane (Ultra residue analysis, J.T. Baker) and then adjusted to a volume of 10 mL.

Mussel and oyster samples were homogenized with an ultra-disperser. Homogenized samples were freeze-dried and seven species of internal standard (ES 2044, Cambridge Isotope Laboratories, Inc.) was spiked. They were decomposed in 200 mL of 1 N KOH ethanolic solution for 2 hours by mechanical shaking. The digest was liquid-liquid extracted with twice using 150 mL of *n*-hexane after addi-

tion 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.

The extracts of sediments and bivalve tissues 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 or two days to evaporate to 100~200 μL. The residues were dissolved with 100 μL of n-nonane (Pesticide residue analysis, Fluka) and determined for PAHs.

The aromatic fraction was analyzed by gas chromatography coupled to mass spectrometery. An Agilent 6890 series II 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 5 ℃/min. The carrier gas was helium at a constant flow rate of 1.2 mL/min. The capillary column used was an DB-5MS (30 m, 0.25 mm ID, 0.25 μm film thickness, J & W Scientific, USA). The GC was coupled to an 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℃. 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 and benzo[g,h,i]perylene (BghiP)). Blanks were run before and after the injection of standard solutions to check for any carryover. Sample recoveries using internal standard were over 85 %. The data presented in this investigation were not corrected for recoveries.

The contents of total organic carbon (TOC) in marine sediment were obtained using a CHN analyzer (Perkin Elmer 2400), after the elimination of the calcium carbonate with 1 N HCl. Grain size analyses were carried out by wet sieving, to separate sands, after a pretreatment with $H_2O_2$. For lipid determination, automatic extraction unit (Gerhardt and Variostat, Germany) was used. About 2 g of freeze-dried bivalve tissue 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.

## Results and Discussion

### Sample characterization

Descriptions for sediment and organism samples were summarized in Table 1. The wide range of sediment contamination levels can be partly explained by the great disparity of the sediments. They ranged from mud to coarse sand, and their organic carbon content ranged from n.d. to 3.25%. In particular, Stations 1, 2 and 3 from East Sea were characterized by primarily sand (94.2~99.4% sand fraction). Wet weight for oysters (C. gigas) and mussels (M. coruscus and M. edulis) was in the range of 0.9~22.5 g. Lipid contents (dry weight basis) in bivalve varied from 4.1 to 10.8%.

### Contaminant levels in sediments

Sixteen PAHs were detected in all sediment samples. Concentrations of ΣPAH (sum of two- to six-ring PAHs) varied from 6.4 to 1,214 ng/g dry weight (Table 2). Station 5 from Pohang coast represents the highest level of ΣPAH. The lowest levels were observed at Stations 1, 2 and 3 from East Sea where grain-size was primarily sandy. This result indicates that grain-size distribution of sediment can be one of important factors governing PAHs concentrations and has to be considered. Indeed, fine grain-size sediments have been shown to accumulate PAHs at greater concentrations than coarse sands (Law and Andrulewicz, 1983). 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]

Table 1.   Sample description of sediments and organisms collected from Korean coast

| Stations | SEDIMENT | | | ORGANISM | | |
| --- | --- | --- | --- | --- | --- | --- |
| | Mud (<63 μm, %) | Sand (>63 μm, %) | TOC (%) | Species | Weight (g) | Lipid (%) |
| 1 | 0.6 | 99.4 | n.d. | Mussel (Mytilus coruscus) | 10.54 (6.0~20.9) | 10.82 |
| 2 | 5.8 | 94.2 | 0.14 | Mussel (Mytilus coruscus) | 2.06 (1.0~3.2) | 8.73 |
| 3 | 0.6 | 99.4 | 0.02 | Mussel (Mytilus coruscus) | 4.61 (2.4~7.8) | 6.22 |
| 4 | 38.2 | 61.8 | 0.59 | Mussel (Mytilus coruscus) | 9.8 (3.7~18.7) | 7.46 |
| 5 | 59.2 | 40.8 | 1.93 | Mussel (Mytilus coruscus) | 8.46 (5.8~13.6) | 10.18 |
| 6 | 94.7 | 5.3 | 1.26 | Mussel (Mytilus coruscus) | 7.16 (3.4~11.9) | 7.24 |
| 7 | 88.8 | 11.2 | 1.47 | Mussel (Mytilus edulis) | 5.02 (4.3~5.8) | 4.14 |
| 8 | 95.4 | 4.6 | 1.65 | Oyster (Crassostrea gigas) | 5.83 (4.4~7.7) | 10.01 |
| 9 | 92.0 | 8.0 | 1.18 | Mussel (Mytilus edulis) | 16.94 (7.3~36.2) | 6.29 |
| 10 | 84.0 | 16.0 | 3.24 | Mussel (Mytilus edulis) | 5.41 (4.5~6.7) | 5.10 |
| 11 | 91.0 | 9.0 | 2.81 | Mussel (Mytilus edulis) | 8.29 (6.7~10.7) | 10.08 |
| 12 | 97.0 | 3.0 | 1.01 | Mussel (Mytilus edulis) | 6.76 (2.7~20.0) | 4.78 |
| 13 | 67.2 | 32.8 | 1.33 | Mussel (Mytilus edulis) | 18.86 (8.8~20.9) | 7.64 |
| 14 | 64.4 | 35.6 | 0.65 | Mussel (Mytilus edulis) | 22.54 (1.0~54.4) | 8.68 |
| 15 | 76.0 | 24.0 | 0.12 | Oyster (Crassostrea gigas) | 3.09 (1.0~8.4) | 9.85 |
| 16 | 63.3 | 36.7 | 0.62 | Oyster (Crassostrea gigas) | 3.2 (1.5~5.4) | 9.23 |
| 17 | 67.6 | 32.4 | 0.51 | Oyster (Crassostrea gigas) | 0.91 (0.5~1.6) | 9.25 |
| 18 | 66.2 | 33.8 | 0.62 | Oyster (Crassostrea gigas) | 5.45 (2.6~10.7) | 8.49 |
| 19 | 54.4 | 45.6 | 0.84 | Oyster (Crassostrea gigas) | 3.89 (2.0~6.9) | 7.75 |
| 20 | 58.1 | 41.9 | 0.76 | Oyster (Crassostrea gigas) | 4.17 (2.0~8.7) | 8.04 |

n.d. = not determined.

Table 2.   The concentration of 16 PAHs in marine sediments from each sampling station of Korean coast

(ng/g dry weight)

| Chemicals | STATIONS | | | | | | | | | | | | | | | | | | | |
| --- | --- | --- | --- | --- | --- | --- | --- | --- | --- | --- | --- | --- | --- | --- | --- | --- | --- | --- | --- | --- |
| | 1 | 2 | 3 | 4 | 5 | 6 | 7 | 8 | 9 | 10 | 11 | 12 | 13 | 14 | 15 | 16 | 17 | 18 | 19 | 20 |
| NaP | 0.27 | 0.21 | 0.77 | 0.89 | 18.26 | 1.77 | 2.58 | 0.28 | 2.24 | 0.70 | 3.58 | 0.29 | 0.27 | 0.84 | 1.30 | 1.91 | 0.23 | 0.31 | 0.26 | 2.05 |
| AcPy | 0.05 | 0.06 | 0.62 | 0.24 | 0.90 | 0.29 | 0.75 | 0.12 | 0.52 | 0.38 | 0.30 | 0.03 | 0.14 | 0.27 | 0.13 | 1.15 | 0.02 | 0.05 | 0.02 | 0.15 |
| AcP | 0.30 | 0.17 | 0.30 | 0.72 | 10.36 | 1.13 | 1.71 | 1.62 | 0.41 | 1.47 | 4.28 | 0.36 | 0.38 | 1.65 | 1.31 | 1.41 | 0.34 | 0.54 | 0.33 | 2.40 |
| Flu | 0.68 | 0.81 | 1.33 | 3.48 | 15.00 | 1.12 | 0.80 | 0.72 | 1.05 | 0.52 | 1.18 | 0.39 | 0.32 | 2.14 | 0.90 | 0.52 | 0.39 | 0.63 | 0.11 | 0.10 |
| - PhA | 0.58 | 0.53 | 1.42 | 4.93 | 107.17 | 12.32 | 10.25 | 2.97 | 10.26 | 5.72 | 7.38 | 2.50 | 1.39 | 2.35 | 3.08 | 1.13 | 0.85 | 2.38 | 1.29 | 1.58 |
| AnT | 0.58 | 0.14 | 0.51 | 0.60 | 17.73 | 2.25 | 1.93 | 0.58 | 1.98 | 1.05 | 1.38 | 0.35 | 0.38 | 3.05 | 3.24 | 0.28 | 0.48 | 0.33 | 0.16 | 0.48 |
| FluA | 0.62 | 2.01 | 0.68 | 11.20 | 146.70 | 35.61 | 23.54 | 9.24 | 20.25 | 20.37 | 18.45 | 15.69 | 6.09 | 3.74 | 6.75 | 3.34 | 3.60 | 12.36 | 7.96 | 7.93 |
| Pyr | 1.90 | 4.89 | 1.37 | 13.93 | 135.26 | 35.11 | 23.48 | 11.42 | 28.24 | 21.28 | 19.13 | 14.65 | 6.02 | 3.98 | 12.24 | 5.94 | 3.99 | 11.70 | 6.20 | 8.10 |
| BaA | 0.17 | 0.20 | 0.20 | 5.71 | 96.05 | 18.05 | 10.55 | 3.62 | 15.21 | 10.52 | 8.45 | 7.57 | 3.03 | 1.17 | 4.99 | 2.06 | 1.10 | 3.18 | 2.09 | 3.20 |
| Chr | 0.16 | 0.52 | 0.19 | 7.08 | 105.25 | 24.59 | 15.23 | 5.32 | 20.21 | 15.47 | 12.13 | 11.95 | 5.82 | 2.71 | 4.64 | 1.92 | 2.33 | 7.03 | 5.26 | 5.70 |
| BbF | 0.22 | 0.61 | 0.25 | 7.82 | 121.10 | 30.28 | 18.35 | 7.47 | 24.17 | 21.90 | 14.73 | 16.23 | 6.43 | 3.82 | 5.84 | 2.40 | 2.59 | 10.08 | 6.91 | 7.15 |
| BkF | 0.18 | 0.52 | 0.17 | 7.86 | 95.20 | 25.60 | 8.95 | 3.33 | 12.25 | 10.65 | 12.20 | 8.18 | 5.27 | 2.58 | 4.18 | 1.84 | 1.90 | 7.28 | 4.82 | 0.30 |
| BaP | 0.21 | 0.60 | 0.12 | 5.11 | 124.94 | 30.62 | 11.93 | 7.18 | 20.58 | 14.20 | 7.50 | 15.29 | 1.73 | 3.46 | 4.90 | 0.68 | 2.99 | 8.20 | 0.63 | 6.73 |
| InP | 0.24 | 0.48 | 0.16 | 7.79 | 109.00 | 26.71 | 20.39 | 7.09 | 23.35 | 25.47 | 11.05 | 14.07 | 6.58 | 2.71 | 4.43 | 1.82 | 1.66 | 8.47 | 4.50 | 4.60 |
| DbA | 0.04 | 0.05 | 0.11 | 0.87 | 16.71 | 4.26 | 2.64 | 0.79 | 3.24 | 3.25 | 1.48 | 1.93 | 0.71 | 0.29 | 0.81 | 0.42 | 0.14 | 0.76 | 0.65 | 0.95 |
| BghiP | 0.26 | 0.44 | 0.30 | 7.13 | 94.55 | 23.58 | 18.26 | 6.44 | 20.98 | 23.77 | 10.90 | 11.60 | 5.94 | 2.69 | 4.10 | 2.18 | 1.73 | 6.97 | 4.11 | 4.48 |
| ΣPAH | 6.4 | 12.3 | 8.5 | 85.3 | 1,214.2 | 273.3 | 171.3 | 68.2 | 204.9 | 176.7 | 134.1 | 121.1 | 50.5 | 37.4 | 62.8 | 29.0 | 24.3 | 80.3 | 45.3 | 55.9 |
| ΣCPAH | 1.1 | 2.5 | 1.0 | 35.2 | 563.0 | 135.5 | 72.8 | 29.5 | 98.8 | 86.0 | 55.4 | 63.3 | 23.7 | 14.0 | 25.1 | 9.2 | 10.4 | 38.0 | 19.6 | 22.9 |

pyrene and dibenzo[a,h]anthracene) (IARC, 1987) varied from 1.0 to 563 ng/g dry weight. The highest concentrations of ΣCPAH were found at Station 5 from Pohang coast. These experimental results were similar to PAH levels in sediments from the south-eastern coastal areas of Korea (Moon et al., 2001).

The PAH levels in sediments measured in this study were summarized in Table 3 with those in other countries. Although the number of PAHs analyzed in other studies may differ, the 16 PAHs

Table 3.  Comparison of PAH concentrations (ng/g dry weight) in sediments measured in this study with those in other countries

| Locations | n | Mean | Range | Reference |
|---|---|---|---|---|
| Korean coast | 16 | 133 | 6.4~1,214 | This study |
| Kyeonggi Bay, Korea | 24 | 120 | 9.1~1,400 | Kim et al. (1999) |
| Casco Bay, USA | 23 | 2,900 | 16~21,000 | Kennicutt et al. (1994) |
| Washington coast, USA | 13 | 200 | 29~460 | Prahl and Carpenter (1983) |
| Chesapeake Bay, USA | 15 | 52 | 0.56~180 | Foster and Wright (1988) |
| San Fansicso Bay, USA | 21 | 8,186 | 2,902~29,590 | Pereira et al. (1996) |
| Manukau Harbour, New Zealnd | 10 | 820 | 16~5,300 | Holland et al. (1993) |
| Humber Plume, North Sea | 13 | 1,800 | 700~2,700 | Klamer and Fomsgaard (1993) |
| Adriatic Sea | 12 | 200 | 18~580 | Caricchia et al. (1993) |
| White Sea | 19 | 74 | 13~208 | Savinov et al. (2000) |
| Baltic Sea | 15 | 700 | 9.5~1,900 | Witt (1995) |
| Western Baltic Sea | 14 | 4,610 | 3~30,000 | Baumard et al. (1999a) |
| Arcachon Bay, France | 14 | 1,918 | 32~4,120 | Baumard et al. (1999b) |
| Gironde Estuary, France | 14 | 256 | 3.5~853 | Soclo et al. (2000) |
| Lazaret Bay, France | 14 | 18,660 | 100~50,000 | Benlahcen et al. (1998) |

n: Number of PAH compounds analyzed in each study.

have found in most estuarine or marine sediments. The PAH concentrations in sediments from Kyeong-gi Bay in Korea were similar or slightly low values compared to these results (Kim et al., 1999). Also, sediment PAH levels measured in this study were comparable to those in Washington coast (Prahl and Carpenter, 1983), Adriatic Sea (Caricchia et al., 1993) and Gironde Estuary, France (Soclo et al., 2000). PAHs levels in sediments from Chesapeake Bay, USA (Foster and Wright, 1988) and White Sea (Savinov et al., 2000) showed lower values than those of Korean coast. However, other reports were about an order of 5~120 times greater than in this study. Consequently, it means that the degree of sediment contamination by PAHs in Korean coast was moderate in comparison to other marine eco-systems.

to PAHs levels in mussels and oysters from Korean coasts measured in this study. However, 14 PAHs levels in mussels (M. galloprovincialis) from Baltic Sea, Arcachon Bay and Mediterranean Sea were in the ranges of 93~361 ng/g dry weight, 279~2,420 ng/g dry weight and 25~20,400 ng/g dry weight, respectively (Baumard et al., 1999b). From these results, the degree of contamination by PAHs in organisms from Korean coast was similar to Baltic Sea and was the lower than those of Arcachon Bay and Mediterranean Sea. Consequently, the degree of organism contamination by PAHs in Korean coast is likely to be moderate. However, periodic monitoring of persistent organic pollutants (POPs) including carcinogenic PAHs should be necessary for a management of environmental quality in Korean coastal ecosystem.

### Contaminant levels in organisms

Sixteen PAHs were detected in all organism samples. Concentrations of ΣPAH varied from 63.2 to 876 ng/g dry weight (Table 4). The highest level was found at Station 8 from Jinhae coast, while the lowest level was Station 4 from Hupo coastal area. The concentrations of ΣCPAH varied from 3.6 to 581.5 ng/g dry weight with the highest concentrations of ΣCPAH at Station 8.

Lee et al. (1998) reported that 16 PAHs concentrations in oysters (C. gigas) collected from Jinhae Bay ranged from 0.1 to 992.0 ng/g. This was similar

### PAHs contributions of sediment and organism

The contributions of ring aromatic groups to sum of 16 PAHs in sediments and organisms from Korean coast were illustrated in Fig. 2. In the sediments, the contribution of 16 PAHs ring groups for each station showed similar patterns for most stations. The predominant contributions were four and five ring aromatics like fluoranthene, pyrene, benzo[a]anthracene, chrysene, benzo[b]fluoranthene, benzo[k]fluoranthene and benzo[a]pyrene, whereas two and three ring aromatics like naphthalene, acenaphthylene, acenaphthene, fluorene, phenanthrene

Table 4.   The concentration of 16 PAHs in organism (mussel and oyster) from Korean coast

(ng/g dry weight)

| Chemicals | STATIONS | | | | | | | | | | | | | | | | | | | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| | 1 | 2 | 3 | 4 | 5 | 6 | 7 | 8 | 9 | 10 | 11 | 12 | 13 | 14 | 15 | 16 | 17 | 18 | 19 | 20 |
| NaP | 6.27 | 5.33 | 6.12 | 5.49 | 6.96 | 5.76 | 7.33 | 6.92 | 7.74 | 5.62 | 5.73 | 5.38 | 6.12 | 10.17 | 10.05 | 6.50 | 6.64 | 7.51 | 6.44 | 6.36 |
| AcPy | 0.62 | 0.02 | 0.50 | 0.00 | 0.41 | 0.47 | 0.41 | 0.39 | 0.76 | 0.28 | 0.44 | 0.00 | 0.38 | 0.53 | 0.54 | 0.77 | 0.50 | 0.45 | 0.40 | 0.51 |
| AcP | 7.69 | 5.93 | 7.00 | 6.28 | 8.94 | 7.42 | 6.79 | 8.51 | 8.21 | 7.43 | 7.31 | 7.37 | 7.38 | 8.25 | 6.03 | 7.80 | 9.66 | 7.08 | 6.37 | 6.97 |
| Flu | 0.68 | 0.71 | 0.71 | 0.82 | 0.96 | 0.81 | 0.61 | 1.49 | 1.77 | 0.63 | 1.43 | 0.82 | 0.89 | 1.69 | 1.51 | 5.33 | 1.66 | 1.08 | 1.11 | 1.10 |
| PhA | 8.45 | 9.88 | 10.14 | 8.48 | 8.56 | 7.96 | 8.17 | 8.74 | 11.63 | 6.13 | 6.68 | 6.51 | 6.36 | 11.99 | 21.17 | 79.64 | 19.86 | 9.02 | 8.52 | 7.62 |
| AnT | 0.21 | 0.78 | 0.39 | 0.30 | 0.40 | 1.72 | 0.42 | 0.56 | 0.57 | 0.14 | 0.48 | 0.23 | 0.37 | 0.38 | 0.92 | 1.63 | 0.25 | 0.43 | 0.83 | 0.36 |
| FluA | 18.89 | 26.06 | 13.42 | 8.62 | 13.04 | 46.36 | 37.99 | 30.66 | 18.29 | 17.63 | 19.77 | 13.48 | 18.60 | 12.69 | 85.62 | 80.00 | 47.74 | 58.09 | 56.55 | 53.20 |
| Pyr | 21.59 | 23.95 | 28.57 | 8.69 | 14.12 | 73.43 | 110.35 | 27.78 | 32.96 | 17.91 | 22.64 | 38.46 | 57.90 | 12.49 | 51.10 | 70.75 | 33.18 | 38.68 | 45.74 | 45.49 |
| BaA | 16.62 | 4.44 | 5.22 | 8.43 | 9.93 | 13.65 | 3.15 | 87.15 | 9.89 | 13.98 | 5.60 | 1.95 | 2.11 | 10.41 | 10.87 | 12.30 | 27.77 | 30.70 | 7.90 | 4.98 |
| Chr | 15.44 | 16.25 | 4.87 | 7.84 | 9.24 | 48.58 | 12.85 | 81.02 | 9.19 | 13.00 | 5.21 | 1.81 | 1.96 | 9.68 | 55.20 | 65.50 | 25.83 | 28.54 | 34.15 | 29.54 |
| BbF | 4.73 | 5.52 | 2.17 | 2.37 | 2.35 | 13.08 | 5.77 | 151.91 | 3.17 | 12.37 | 1.15 | 0.48 | 0.63 | 3.31 | 23.38 | 25.93 | 9.84 | 8.64 | 16.81 | 10.94 |
| BkF | 3.20 | 5.25 | 1.63 | 1.70 | 0.85 | 8.86 | 4.13 | 144.66 | 3.41 | 9.19 | 0.76 | 0.37 | 0.55 | 1.87 | 11.39 | 13.08 | 4.24 | 3.45 | 8.20 | 4.27 |
| BaP | 5.42 | 6.36 | 2.54 | 2.79 | 3.69 | 23.4 | 57.50 | 42.81 | 4.95 | 2.92 | 1.78 | 0.60 | 0.31 | 5.05 | 2.65 | 3.23 | 8.10 | 7.80 | 5.41 | 9.04 |
| InP | 1.45 | 2.94 | 1.23 | 0.55 | 0.91 | 2.51 | 2.31 | 141.21 | 1.55 | 9.26 | 0.36 | 0.17 | 0.30 | 0.68 | 2.52 | 3.76 | 1.27 | 0.75 | 3.88 | 0.96 |
| DbA | 0.22 | 0.37 | 0.15 | 0.19 | 0.13 | 0.80 | 0.33 | 13.76 | 0.25 | 0.91 | 0.12 | 0.05 | 0.10 | 0.18 | 0.68 | 0.64 | 0.29 | 0.16 | 0.55 | 0.23 |
| BghiP | 1.90 | 2.43 | 1.37 | 0.66 | 1.16 | 3.83 | 2.89 | 128.50 | 1.69 | 9.15 | 0.54 | 0.26 | 0.35 | 1.31 | 2.69 | 4.71 | 1.76 | 0.92 | 4.01 | 1.25 |
| ΣPAH | 113.3 | 116.2 | 86.0 | 63.2 | 81.6 | 258.6 | 210.9 | 876.0 | 116.0 | 126.5 | 79.9 | 77.9 | 104.3 | 90.7 | 286.3 | 381.5 | 198.6 | 203.3 | 206.8 | 182.8 |
| ΣCPAH | 31.6 | 24.9 | 12.9 | 16.0 | 17.9 | 62.3 | 23.2 | 581.5 | 23.2 | 48.6 | 9.8 | 3.6 | 4.0 | 21.5 | 51.5 | 58.9 | 51.5 | 51.5 | 42.7 | 30.4 |
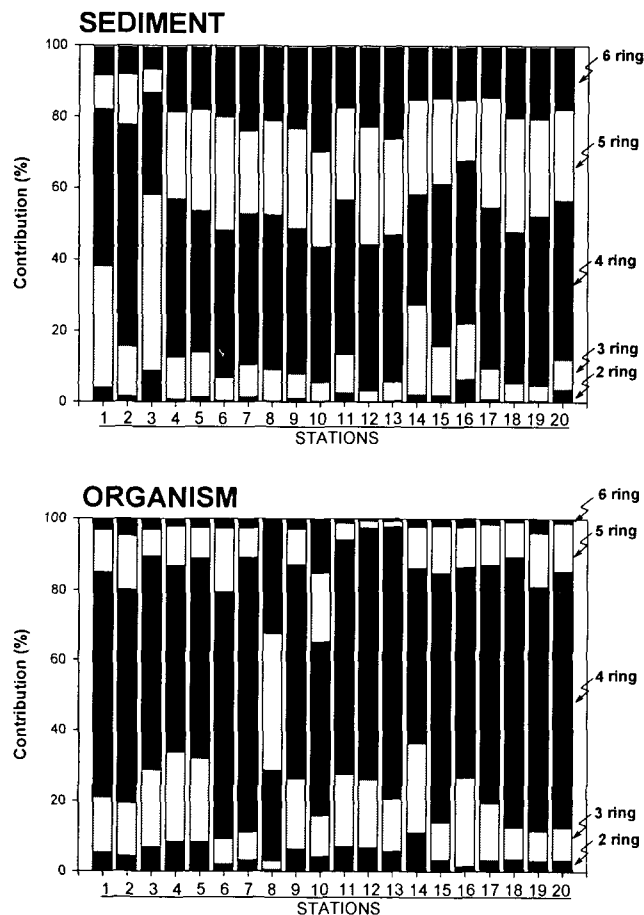


Fig. 2.   Contributions of ring aromatic groups (two to six ring) to total concentrations of 16 PAHs in marine sediments and organism from Korean coast.

and anthracene showed a low concentration. The higher-molecular-weight four and five ring aromatics with more lipophilic and hydrophobic character predominated in the sediments matrix, probably due to their higher persistence. These patterns were in accordance with a typical profile of PAHs in marine sediments and particulate matter in ambient air from previously published references (Witt, 1995; Benlahcen et al., 1998; Ok et al., 1998). These results indicate that the higher-molecular-weight aromatics seem to be preferentially adsorbed to particles and incorporated in the sediment. However, Stations 1, 2 and 3 from East Sea showed different contributions. PAHs contribution in sandy sediments from Stations 1, 2 and 3 was dominated by three and four ring aromatics, because persistent ability of higher-molecular-weight PAHs to sandy sediments was relatively lower than muddy sediments.

In the organisms, the contribution of four ring aromatics like fluoranthene, pyrene, benzo[a]anthracene and chrysene ranged between 50 and 77% in exception with Station 8. In particular, Station 8 from Jinhae coast with a high percentage of higher-weight-molecular five and six ring aromatics was characterized by the highest concentration in this study. The results suggest that the contamination of PAHs might be influenced by combustion process of high temperature and a local point source also

might give the higher burden on PAHs contamination in the marine environment.

## Source of PAHs contamination in the studied stations

The sources of PAH contaminations, whether from combustion process of fuels (pyrolytic origin) or from oil contamination (petrogenic origin), may be identified by ratios of individual PAH compound based on characteristics in PAH composition. The ratio of phenanthrene/anthracene and fluoranthene/pyrene allows us to give useful information on source of PAH contamination (Mille et al., 1982). Hence, phenanthrene/anthracene<10 and fluoranthene/pyrene>1 were mainly contaminated by petrogenic inputs and phenanthrene/anthracene>10 and fluoranthene/pyrene<1 was typical of pyrolytic origin (Colombo et al., 1989; Garrigues et al., 1995). Fig. 3 clearly shows that the ratios of phenanthrene /anthracene were lower than 10 (values between 0.95 to 8.21) for all stations. According to this result, PAHs contamination for all stations was influenced by pyrolytic origin. The ratios of fluoranthene/pyrene varied from 0.33 to 1.28. Most stations showed that the ratios of fluoranthene/pyrene were lower than 1. The results indicate that contamina-



Fig. 3. The plots between the ratio of phenanthrene/anthracene and fluoranthene/pyrene in sediments form Korean coast.

tion by PAHs of these coasts is based on petrogenic origin. However, the ratio values of Stations 5, 6, 12, 13 and 19 were higher than 1 (values between 1.01 and 1.28), showing pyrolytic origin. Indeed, Stations 5 (Pohang coast), 6 (Ulsan coast), 12 (Gwangyang coast), and 13 (Yeosu coast) were located close to a large number of industrial complex in Korea, so these stations clearly showed pyrolytic origin of PAHs from various combustion processes.

Soclo et al. (2000) investigated the origin index of the PAH contamination using the ratio of the sum of low molecular weight (LMW; groups with three and four aromatic rings) versus the sum of high molecular weight (HMW; groups with five and six aromatic rings). The choice of this origin index was founded on the fact that the petrogenic contamination was characterized by the predominance of the lower molecular weight PAHs (three and four ring aromatics) (Neff, 1979; Berner et al., 1990), while the higher molecular weight PAHs dominated in the pyrolytic PAH contamination distribution (Muel and Saguem, 1985). In most stations, the ratio values of LMW/HMW showed lower than 1 being in the range of 0.38 to 0.80. Only, LMW/HMW ratio values in sediment samples from Stations 1, 2 and 3 revealed higher than 1 ranging from 2.21 to 2.65. The difference of ratio among stations resulted from the sediment characters such as grain-size. Therefore, it seems that the majority of these sampling stations is characteristic of mainly pyrolytic origin with slight petrogenic input.
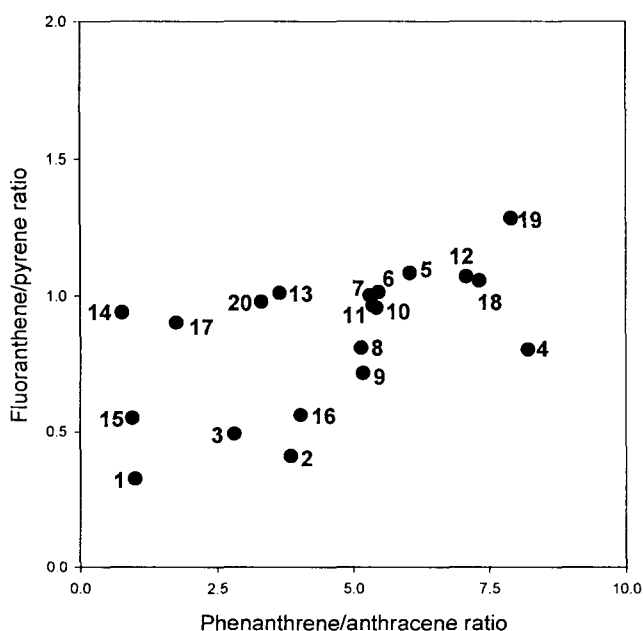
## Correlation analysis

Correlation factors between individual PAH compound in sediment and organism samples from the coastal areas of Korea were summarized in Table 5. In the sediments, there was a highly positive correlation between the concentrations of PAH species with the exception of acenaphthylene ($r=0.43\sim0.54$, $0.01<p<0.05$, $n=20$). In particular, correlation factors among high-molecular-weight five and six ring aromatics showed higher than those among low-molecular-weight two and three ring aromatics.

In the organisms, there was no correlation within lower molecular weight PAHs unlike marine sediments. However, the relationships within four to six ring aromatic PAHs, namely benzo[a]anthracene, chrysene, benzo[b]fluoranthene, benzo[k]fluoran-

Table 5.   Pearson product-moment correlation coefficients for 16 PAHs in organism and sediment

| | NaP | AcPy | AcP | Flu | PhA | AnT | FluA | Pyr | BaA | Chr | BbF | BkF | BaP | InP | DbA | BghiP |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| | | | | | | | ORGANISM | | | | | | | | | |
| NaP | | 0.481* | 0.121 | 0.193 | 0.114 | 0.038 | 0.322 | 0.054 | 0.112 | 0.217 | 0.090 | 0.053 | 0.002 | 0.026 | 0.038 | 0.028 |
| AcPy | 0.540* | | 0.401 | 0.533* | 0.454* | 0.319 | 0.415 | 0.258 | 0.102 | 0.330 | 0.058 | 0.010 | 0.058 | -0.028 | -0.015 | -0.021 |
| AcP | 0.955*** | 0.473* | | 0.255 | 0.131 | -0.159 | -0.197 | -0.203 | 0.412 | 0.061 | 0.223 | 0.240 | 0.263 | 0.258 | 0.251 | 0.262 |
| Flu | 0.949*** | 0.445* | 0.884*** | | 0.953*** | 0.554* | 0.516* | 0.202 | 0.103 | 0.506* | 0.173 | 0.105 | -0.023 | 0.051 | 0.064 | 0.058 |
| PhA | 0.983*** | 0.471* | 0.914*** | 0.969*** | | 0.590** | 0.570** | 0.291 | 0.002 | 0.492* | 0.081 | 0.005 | -0.107 | -0.054 | -0.039 | -0.047 |
| AnT | 0.966*** | 0.451* | 0.909*** | 0.960*** | 0.978*** | | 0.605** | 0.485* | 0.000 | 0.636** | 0.141 | 0.077 | 0.261 | 0.006 | 0.043 | 0.015 |
| FluA | 0.964*** | 0.450* | 0.902*** | 0.935*** | 0.987*** | 0.955*** | | 0.537* | 0.107 | 0.717*** | 0.167 | 0.059 | 0.111 | -0.019 | 0.011 | -0.013 |
| Pyr | 0.962*** | 0.466* | 0.894*** | 0.930*** | 0.984*** | 0.955*** | 0.997*** | | -0.157 | 0.294 | -0.016 | -0.058 | 0.095 | -0.100 | -0.079 | -0.093 |
| BaA | 0.973*** | 0.467* | 0.900*** | 0.948*** | 0.994*** | 0.968*** | 0.996*** | 0.996*** | | 0.656** | 0.916*** | 0.918*** | 0.852*** | 0.915*** | 0.917*** | 0.916*** |
| Chr | 0.962*** | 0.455* | 0.891*** | 0.930*** | 0.985*** | 0.954*** | 0.998*** | 0.997*** | 0.997*** | | 0.749*** | 0.676*** | 0.689*** | 0.611** | 0.640** | 0.618** |
| BbF | 0.953*** | 0.449* | 0.885*** | 0.919*** | 0.979*** | 0.947*** | 0.997*** | 0.996*** | 0.994*** | 0.999*** | | 0.993*** | 0.866*** | 0.981*** | 0.986*** | 0.982*** |
| BkF | 0.957*** | 0.435 | 0.890*** | 0.941*** | 0.984*** | 0.955*** | 0.995*** | 0.992*** | 0.993*** | 0.993*** | 0.991*** | | 0.874*** | 0.996*** | 0.999*** | 0.997*** |
| BaP | 0.955*** | 0.433 | 0.880*** | 0.933*** | 0.984*** | 0.957*** | 0.994*** | 0.993*** | 0.995*** | 0.995*** | 0.994*** | 0.991*** | | 0.863*** | 0.876*** | 0.867*** |
| InP | 0.940*** | 0.467* | 0.869*** | 0.908*** | 0.971*** | 0.936*** | 0.991*** | 0.993*** | 0.988*** | 0.995*** | 0.997*** | 0.983*** | 0.988*** | | 0.999*** | 0.999*** |
| DbA | 0.951*** | 0.473* | 0.878*** | 0.915*** | 0.977*** | 0.947*** | 0.994*** | 0.995*** | 0.993*** | 0.997*** | 0.998*** | 0.988*** | 0.993*** | 0.998*** | | 0.999*** |
| BghiP | 0.939*** | 0.474* | 0.872*** | 0.904*** | 0.968*** | 0.934*** | 0.989*** | 0.992*** | 0.986*** | 0.993*** | 0.996*** | 0.981*** | 0.985*** | 0.999*** | 0.997*** | |
| | | | | | | | SEDIMENT | | | | | | | | | |

*$0.01<p<0.05$, **$0.001<p<0.01$, ***$p<0.001$.

thene, benzo<a>pyrene, indeno<1,2,3-c,d>pyrene, dibenzo<a,h>anthracene and benzo<g,h,i>perylene, showed a significant correlation. This finding is thought to be relation with a metabolism of organisms. In the marine environment, the majority of heavier molecular weight PAHs was strongly adsorbed with particulate matter because of hydrophobic and persistent character of PAHs. Filter-feeding marine bivalves such as oysters and mussels can absorb xenobiotics adsorbed on a small grain-size fraction of particles through digestive system. PAHs migrated with particle into organism are accumulated to a greater extent. Therefore, heavier molecular weight PAHs in organism matrix is thought to be a high correlation relative to low molecular weight PAHs.

## Conclusion

Sediments and organisms have been sampled in several areas from Korean coast to investigate levels and sources of PAH contamination. PAHs levels in sediment and organism were low or moderate comparable to other urbanized countries of the world. The contribution of ring aromatic groups to sum of 16 PAHs in sediments and organisms showed similar patterns for most stations. Several molecular indices such as phenanthrene/anthracene, fluoran-

thene/pyrene and LMW/HMW ratio were shown to be useful to differentiate between the different contamination sources. In this study, PAHs are mainly of pyrolytic origin with slight petrogenic input. A high correlation within individual PAH compound in marine sediments was observed, while correlation in organisms showed a highly significant relationship for heavier molecular weight PAHs.

## References

Baumard, P., H. Budzinski, P. Garrigues, H. Dizer and P.D. Hansen. 1999a. 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.

Baumard, P., H. Budzinski, P. Garrigues, J.F. Narbonne, T. Burgeot, X. Michel and J. Bellocq. 1999b. Polycyclic aromatic hydrocarbon (PAH) burden of mussels (Mytilus sp.) in different marine environments in relation with sediment PAH contamination and bioavailability. Mar. Environ. Res., 47, 415~439.

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

Berner, B.A., N.P. Bryner, S.A. Wise, G.H. Mulholland, R.C. Lao and M.F. Fingas. 1990. Polycyclic aromatic hydrocarbon emissions from combustion crude oil on water. Environ. Sci. Technol., 24, 1418~1427.

Caricchia, A.M., S. Chiavarini, C. Cremisini, F. Marrini and

R. Morabito. 1993. PAHs, PCBs, and DDE in the nor-
thern Adriatic Sea. Mar. Pollut. Bull., 26, 581~583.

Claisse, D., M. Joanny and J.Y. Quintin. 1992. Le reseau
national d'observation de la qualite du milieu marin
(RNO). Analysis, 20, 19~22 (in French).

Colombo, J.C., E. Pelletier, C. Brochu, M. Khalil and J.A.
Catoggio. 1989. Determination of hydrocarbon courses
using n-alkane and polynuclear aromatic hydrocarbon
distribution indexs. Case study: Rio de La Plata Estuary,
Argentina. Environ. Sci. Technol., 23, 888~894.

Farrington, J.W., E.D. Goldberg, R.W. Risegrough, J.H. Martin
and V.T. Bowen. 1983. An overview of the trace-metal.
DDE, PCB, hydrocarbon, and artificial radionuclide data.
Environ. Sci. Technol., 17, 490~496.

Foster, G. and D.A. Wright. 1988. Unsubstituted polynuclear
aromatic hydrocarbons in sediments, clam worms from
Chesapeake Bay. Mar. Pollut. Bull., 19, 459~465.

Garrigues, P., H. Budzinski, M.P. Manitz and S.A. Wise. 1995.
Pyrolytic and petrogenic inputs in recent sediments: a
definitive signature through phenanthrene and chrysene
distribution. Pol. Arom. Comp., 7, 275~284.

Goldberg, E.D., V.T. Bowen, J.W. Farrington, G. Harvey, J.H.
Martin, P.L. Partker, R.W. Riesebrough, W. Robertson, E.
Schneider and E. Gamble. 1978. The mussel watch.
Environ. Conserv., 5, 101~125.

Hites, R.A., R.E. LaFlamme and J.W. Farrington. 1977. Sedi-
mentary polycyclic aromatic hydrocarbons: The historical
report. Science, 198, 829~831.

Holland, P.T., C.W. Hickey, D.S. Roper and T.M. Trower.
1993. Variability of organic contaminants in inter-tidal
sandflat sediments from Manukau Harbour, New Zea-
land. Arch. Environ. Contam. Toxicol., 25, 456~463.

IARC, 1987. IARC monographs on the evaluation of the car-
cinogenic risk of chemicals to humans, overall evaluation
of carcinogenicity: an updating of IAPC monographs
volumes 1~42, Suppl. International Agency for Research
on Cancer, Lyon, France, pp. 1~477.

Kennicutt, M.C., T.L. Wade, B.J. Presley, A.G. Requejo, J.M.
Brooks and G.J. Denoux. 1994. Sediment contaminants
in Casco Bay, Marine: Inventories, sources, and potential
for biological impact. Environ. Sci. Technol., 28, 1~15.

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.

Klamer, H.J.C. and L. Fomsgaard. 1993. Geographical distri-
bution of chlorinated biphenyls (CBs) and polycyclic
aromatic hydrocarbons (PAHs) in surface sediments
from the Humber Plume, North Sea. Mar. Pollut. Bull.,
26, 201~206.

Law, R.J. and E. Andrulewicz. 1983. Hydrocarbons in water,
sediment and mussels from the southern Baltic Sea. Mar.
Pollut. Bull., 14, 289~293.

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

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.

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.

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

Mille, G., J.Y. Chen and H.J.M. Dou. 1982. Polycyclic aroma-
tic hydrocarbons in mediterranean coastal sediments.
Inter. J. Environ. Anal. Chem., 11, 295~304.

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. 2001. Polycyclic aromatic hydrocarbons in sediments
from the southeastern coastal areas of Korea. J. Kor. Soc.
Oceanogr., 36, 27~33.

Muel, B. and S. Saguem. 1985. Determination of 23 polycyclic
aromatic hydrocarbons in atmospheric particulate matter
of the Paris area and photolysis by sun light. Inter. J.
Environ. Anal. Chem., 19, 111~131.

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

O'Connor, T.P. 1996. Trends in chemical concentrations in
mussels and oysters collected along the coasts from 1986
to 1993. Mar. Environ. Res., 41, 183~200.

Ok, G., H.B. Moon, S.H. Ji and Y.H. Han. 1998. Concent-
ration levels and characteristics of polycyclic aromatic
hydrocarbons (PAHs) in atmospheric suspended parti-
culate matters. J. Kor. Soc. Environ. Anal., 1, 265~274
(in Korean).

Oliver, B.G. and A.J. Niimi. 1988. Trophodynamic analysis of
polychlorinated biphenyl congeners and other chlorina-
ted hydrocarbons in the Lake Ontario ecosystem. Envi-
ron. Sci. Technol., 22, 388~397.

Pereira, W.E., F.D. Hostettler and J.B. Rappe. 1996. Distribu-
tion and fate of chlorinated pesticides, biomarker and
polycyclic aromatic hydrocarbons in sediments along con-
tamination gradient from a point-source in San Fran-
sicso Bay, California. Mar. Environ. Res., 41, 299~314.

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 analy-
sis. Mar. Pollut. Bull., 34, 85~95.

Phillips, D.J.H. 1978. Use of biological indicator organisms to
quantitative organochlorine pollutants in aquatic en-
vironments: A review. Environ. Pollut., 16, 167~229.

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

Readman, J.W., R.F.C. Mantoura, M.M. Rhead and L. Brown.
1982. Aquatic distribution and heterotrphic degradation
of polycyclic aromatic hydrocarbons (PAHs) in the

Tamar Estuary. Est. Coast. Shelf Sci., 14, 369~389.

Savinov, V.M., T.N. Savinova, J. Carroll, G.G. Matishov, S. Dahle and K. Naes. 2000. Polycyclic aromatic hydrocarbons (PAHs) in sediments of the White Sea, Russia. Mar. Pollut. Bull., 40, 807~818.

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.

Spies, R.B., D.W. Rice and J. Felton. 1988. Effects of organic contaminants on reproduction of the starry flounder *Platichthys stellatus* in San Francisco Bay. I. Hepatic contamination and mixed-function oxidase (MFO) activity during the reproductive season. Mar. Biol. 98, 181~189.

Wakeham, S.G., C. Schaffner and W. Giger. 1980. Polycyclic aromatic hydrocarbons in recentlake 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.