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論 文

Polycyclic Aromatic Hydrocarbons(PAHs) in Sediment and Mussels(*Mytilus edulis*) from the Intertidal Zone of Kori Nuclear Power Plant, Korea

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고리원자력발전소 인근 조간대에 서식하는 퇴적물과 진주담치에 포함된 다환방향족 탄화수소(PAHs)

노 일 · 이 기 석

〈목 차〉

Abstract

Polycyclic aromatic hydrocarbons (PAHs) are ubiquitous contaminants in coastal marine environment. PAHs enter estuarine and nearshore marine environment via several routes such as combustion of fossil fuels, domestic and industrial effluents and oil spills.

In August of 1997, sediment and mussels (*Mytilus edulis*) were collected at 6 sites near Kori nuclear power plant in order to analyze the PAH content by HPLC with uv/vis detection.

The concentrations of 15 PAH in sediment ranged from < 1 to 5,900 ppb (mean 173.5±99.7 ppb), and in mussels, from < 0.5 to 4,125 ppb (mean 105±60.5 ppb).

Compared with other studies world over, the concentrations of carcinogenic PAHs were relatively low in both sediment and mussels from the intertidal zone of Kori.

This study presents preliminary data for the PAH levels in sediment and mussels from the intertidal zone of Kori, and the data will hopefully be utilized for the assessment of oil pollution in the Southeast East Sea, Korea (especially for the PAHs).

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INTRODUCTION

Estuarine and coastal environments are apt to be affected by various kinds of pollutants. Of all things, oil pollution have recently received the greatest public attention because of the direct damage to fisheries, and the harmful effects on marine lives.

Polycyclic or polynuclear aromatic hydrocarbons (PAHs) are a group of compounds composed of two or more fused aromatic rings. PAHs have been the focus of numerous studies in the world because they are potentially carcinogenic, mutagenic, and teratogenic to aquatic organisms and humans consuming PAH-contaminated food.

PAHs enter marine environment via several routes; domestic and industrial effluents, oil spill, incomplete combustion of fossil fuels, forest and brush fires, terrestrial contributions and natural sources such as biosynthesis by plant and microorganisms. However, oil spill and incomplete combustion of fossil fuels are major sources of PAHs in marine environment (Neff, 1985; Rainio *et al.*, 1986). They typically adsorb to fine particulate material suspended in estuarine waters and sediment seafloor (Law and Whinett, 1992).

Once PAHs have been introduced to a harbor or coastal zone, they accumulate in sediments because of their hydrophobicity and partitioning to the organic carbon-coated particles (Means *et al.*, 1980). Since PAHs have low solubility in water and tend to be transported with suspended sediment, most PAHs introduced into aquatic environments accumulate in bottom sediment. PAHs, as well as other organic pollutants, remain in the sediment usually depending on their rate of degradation in sediments (Delanne *et al*, 1981). Mussels are intertidal and subtidal organisms, which attach themselves to various substances and filter-feed on suspension.

No measurement of PAHs in sediment and mussels taken from the Southeast Sea of Korea has been reported up to the present.

This study presents preliminary data on the PAH levels in sediment and mussels from the intertidal zone near Kori nuclear power plant. Moreover, we tried to trace the sources of PAHs in sediment and mussels. And this study will contribute to the establishment of database on PAH contamination in sediment and mussels in the Southeast Sea of Korea.

MATERIALS AND METHODS

Chemicals

15 unsubstituted PAHs in coastal environment near Kori nuclear power plant were investigated: Naphthalene (NPTHL), Acenaphthylene (ANCPL), Acenaphthene (ACNPN), Fluorene (FLURN), Phenanthrene (PHEN), Anthracene (ANTHR), Fluoranthene (FLRTH), Pyrene (PYR), Chrysene (CHRY), Benzo(b)fluoranthene (BbF), Benzo(k) fluoranthene (BkF), Benzo(a)pyrene (BaP), Dibenz(a,h)anthracene (DahA), Benzo(g.h.i) peryline (BghiP) and Indeno(1,2,3,-cd)pyrene (I123cdP). Of those, EPA recently reported that the following seven PAHs are typically considered as possible or probable carcinogens: Chrysene (CHRY), Benzo(b)fluoranthene (BbF), Benzo(k)fluoranthene (BkF), Benzo(a)pyrene (BaP), Dibenz(a,h)anthracene (DahA), Benzo (g.h.i)peryline (BghiP) and Indeno(1,2,3,-cd) pyrene (I123cdP) (Menzie *et al.*, 1992).

Sampling and Extraction of PAHs from mussels (*Mytilus edulis*) and sediment

*M*ussels (*Mytilus edulis*) and surface sed-

iment were collected at six sites by SCUBA divers from 1.5~3 m water depth near the intertidal zone of Kori nuclear power plant, Korea, during August of 1997 (Fig. 1). The shell length of the mussels ranged from 3cm to 3.8cm, indicating young specimens. Samples were stored frozen under darkness in an icebox to minimize possible degradation caused by photo-oxidation or bacterial action and brought to the lab, being stored at 20℃ prior to analysis. Sampling device was cleaned with distilled water in between each collection.

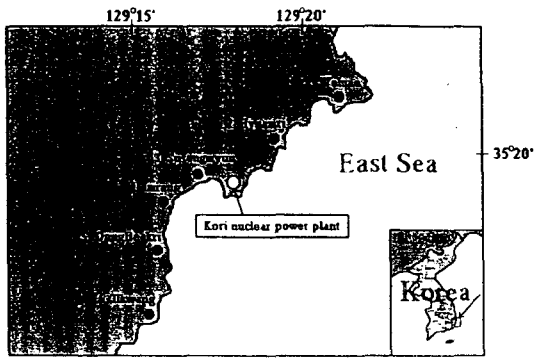

Fig. 1. Location of sampling sites in the Southeast Sea, Korea

Sampling sites were located in near residential area. Domestic wastewater and runoff have been discharged into this area via several streams. In summer seasons, red tides tend to occur annually in this area.

Glass devices were prepared by heating at 450℃ to remove any possible organic contamination.

The extraction method used in this study has been described in Lee (1997). HPLC grade reagents (hexane, acetone, diethylether, petroleum ether, methanol, dimethylsulfoxide (DMSO), cyclohexane, etc.) and ultrapure distilled water were used for all extraction procedures. The shells were removed and the tissue of mussels (20g, wet wt) were homogenized with macerator and dried with anhydrous sodium sulfate (Na_2SO_4). The fats in thimble filter were Soxhlet-extracted with a mixture of hexane, acetone, diethylether and petroleum ether for 6 hour (Rainio *et al.*, 1986). Then the fatty residues mixed with methanol containing 7g of potassium hydroxide (KOH) were refluxed for 3 hours. After digestion, the solution was separated three times with a cyclohexane (Smith *et al.*, 1984; Smith *et al.*, 1987). To separate PAHs from the aliphatic hydrocarbons, the liquid-liquid extraction procedure developed by Natusch and Tomkins(1978) was applied. The dimethylsulfoxide (DMSO) layers, which contained the PAHs, were then combined. For cleanup of PAHs, two volumes of water were added to the combined DMSO extracts. The resulting solution was partitioned three times with equal volume of cyclohexane. The cyclohexane layers were washed once with equal amount of water(Rainio, 1986), and were dried nearly to a volume of 1.5mℓ with a rotary evaporator at 40℃, and then the sample was filtered through a 0.45㎛ PVDF filter(Whatman). Finally the samples were concentrated to a final volume of approximately 1mℓ under a stream of nitrogen gas, and the sample vials were stored in the freezer prior to analysis by HPLC.

The 20g of sediment samples was thawed and excess water was removed through centrifugation and air-dried in the dark. Sediment samples with organic materials were ground by mortar and pestle and sieved with 0.5mm sieve. Subsequent extraction procedure for the PAH analysis of sediment was done according to the procedure for the extraction of PAHs from mussels.

Concentrations of PAHs in sediment are expressed in units of ㎍/㎏ dry weight.

HPLC system

The analysis of PAHs from sediment and mussels were carried out by computer-assisted HPLC system (Linear Instrument Co.), equipped with a Model S-1100 binary solvent delivery system, a Model S-2000 automatic gradient controller and an injector fitted with a GROM-SIL 120 ODS-5 ST column (250×4㎜, 5 ㎛ particle size).

The flow rate of mobile phase was held constant at 0.8 ㎖/min under the condition of 0.5 bar pressure.

Solvent A of acetonitril, and solvent B composed of ultrapure distilled water and acetonitril (50 : 50, v/v), were utilized as mobile phases.

A gradient solvent system for the elution of PAHs in this study was programmed as follows: delivery program of solvent was planned at 100 % solvent B for the initial condition, 70 % solvent B at 5 min, 20 % solvent B at 15 min, 10 % solvent B at 20 min, 5 % solvent B at 25 min, solvent A 100% at 30 min, followed by an isocratic hold until all PAHs peaks were eluted. (Table 1).

Table 1. Binary gradient program used in this study

Time (min)	Solvent A CH_3CN (%)	Solvent B H_2O : CH_3CN (50 : 50, v/v) (%)
0	0	100
5	30	70
15	80	20
20	90	10
25	95	5
30	100	0

Sample was injected using 20㎕ syringe. Analytical blank test was carried out between each sample run and no analytical contamination has found in the HPLC system.

The peaks of PAHs were identified and quantified using uv/vis detection setting at 254 nm.

The management of chromatograms, integration and calibration of data were carried out using Peaksimple Serial Data Program system (SRI Model 202).

RESULTS AND DISCUSSION

Dominant PAH compounds

The chemical formula, structure, and retention times of each compound of PAHs are summarized in Table 2.

In the present study, certified reference material was used for verification of PAHs, and each PAH was identified on the basis of retention time marked in the chromatograms and quantified by uv/vis detector responses of the samples with the corresponding peaks of authentic standard solution (Supelco: Lot 125H0792). Separation for individual NPTHL, ANCPL, PHEN, ANTHR, FLRTH, PYR, CHRY, BaP, DahA, BghiP and I123cdP in the standard solution was good enough by the HPLC system, while ACNPN, FLURN, BbF and BkF were not sharply separated with uv/vis detection (Fig. 2).

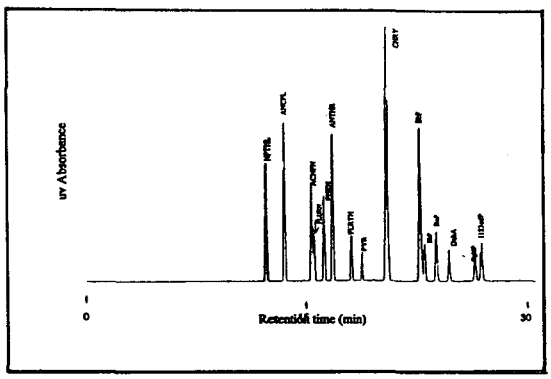

Fig. 2 Chromatogram of PAH standard solution by HPLC with uv/vis detection. BaA can't be eluted set at 254nm.

Table 2. The chemical formula, structures and retention times (R*t*) of PAHs analyzed in the study

mw: molecular weight

No.	COMPOUND (ABBREV)	ALTERNATIVE NAME	FORMULAR (MW)	STRUCTURE	R*t* (min)*
1	Naphthalene(NPTHL)		$C_{10}H_8$ (128)		12.16
2	Acenaphthylene(ANCPL)		$C_{12}H_8$ (152)		13.28
3	Acenaphthene(ACNPN)		$C_{12}H_{10}$ (154)		15.15
4	Fluorene(FLURN)		$C_{12}H_{10}$ (166)		15.38
5	Phenanthrene(PHEN)		$C_{14}H_{10}$ (178)		16.00
6	Anthracene(ANTHR)		$C_{14}H_{10}$ (178)		16.55
7	Fluoranthene(FLRTH)		$C_{16}H_{10}$ (202)		17.80
8	Pyrene(PYR)		$C_{16}H_{10}$ (202)		18.55
9	Chrysene(CHRY)		$C_{18}H_{12}$ (228)		20.13
10	Benzo(b)fluoranthene(BbF)	3,4 Benzfluoranthene	$C_{20}H_{12}$ (252)		22.40
11	Benzo(k)fluoranthene(BkF)	11,12 Benzfluoranthene	$C_{20}H_{12}$ (252)		22.73
12	Benzo(a)pyrene(BaP)	3,4 Benzopyrene	$C_{20}H_{12}$ (252)		23.45
13	Dibenz(a.h)anthracene(DahA)	1,2,5,6 Dibenzanthracene	$C_{22}H_{14}$ (278)		24.33
14	Benzo(g.h.i)peryline(BghiP)	1,12 benzperylene	$C_{22}H_{12}$ (276)		26.06
15	Indeno(1.2.3-cd)pyrene ($I_{123\ cd}P$)	o-Phenylenepyrene	$C_{22}H_{12}$ (276)		26.40

* The retention times of PAHs analyzed were drawn from uv/vis detection.

The concentrations of PAHs in mussels and sediment for six sites are presented in Table 3 and 4. And the representative chromatograms of PAHs of mussels and sediment analyzed by uv/vis detection at site 6 are presented in Fig. 3. The highest PAH in sediment was ACNPN, while in mussels it was ACNPL. I123cdP was under detection limit in mussels from all sites. Moreover, that PAH compound was almost under detection limit in sediment only except at site 5 and 6.

Mean Concentration

The mean concentration of each PAH compound in sediment ranged from 1.8 to 1,090ppb, and that in mussel ranged from 3.2 to 1,068ppb. The mean concentration of each

Table 3. The concentration of PAHs in sediment (μg/kg dry wt) from the intertidal zone of Kori.

	Site 1	Site 2	Site 3	Site 4	Site 5	Site 6	Mean±SE*
NPTHL	237.5	33	38.5	193	55.5	800	226±97
ANCPL	ND	22.5	66	51.5	240	365	149±65
ACNPN	62	ND	4	172.5	ND	4,125	1,090±1011
FLURN	246	4	ND	ND	35	18.5	75±57
PHEN	7.5	3	3	4	5	6.5	4.8±0.7
ANTHR	1.5	27.5	24.5	ND	ND	23	19±1
FLRTH	85.5	2.5	2.5	ND	25	108	44±22
PYR	43	35.5	19.5	6	ND	122	45±20
CHRY	2	1	1	0.5	1	2	1.3±0.3
BbF	1.5	2	ND	2	2	ND	1.9±0.1
BkF	1.5	ND	2.5	ND	ND	11	11.7±10
BaP	ND	1	ND	3	3	ND	2.3±0.6
DahA	ND	4.5	ND	ND	6.5	25	12±5
BghiP	ND	ND	ND	5.5	ND	ND	5.5
$I_{123cd}P$	ND	ND	ND	ND	2	1.5	18±0.2
Total	688	136.5	161.5	438	375	5607.5	1,234±878

*SE : Standard Error, ND: Not detected

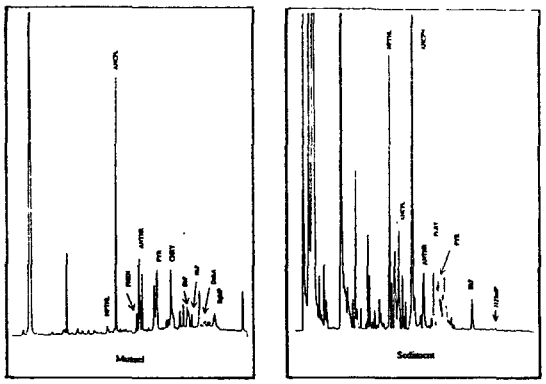

Fig. 3. Chromatograms of PAHs in mussels and sediment with uv/vis detection at site 6.

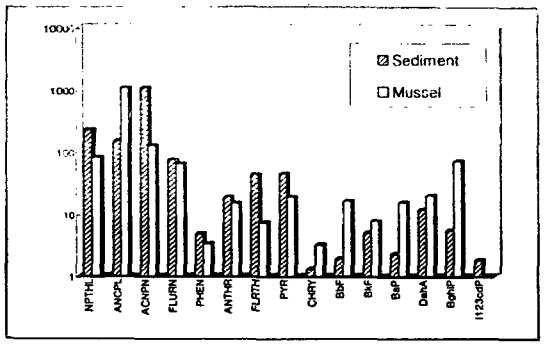

Fig. 4. The mean concentration of each PAH compound in sediment versus messels

PAH compound in sediment versus mussel is shown in Fig. 4. The mean concentrations of NPTHL, ACNPN, FLURN, PHEN, ANTHR, FLRTH, and PYR in sediment were higher than those in mussel, and those of the rest of PAHs in sediment, which are mostly higher molecular PAHs, were much lower than those in mussels. The reason for that is not clear yet. It may be presumed that the higher the molecular weight of PAHs are, the easier they can be attacked by microorganisms in sediment than they are *in*

Table 4. The concentration of PAHs in mussels (μg/kg wet wt) from the intertidal zone of Kori.

	Site 1	Site 2	Site 3	Site 4	Site 5	Site 6	Mean±SE*
NPTHL	22.5	85.5	334	16.5	25	2.5	81±51
ANCPL	42	5,900	3,260	290	26.5	565	1,068±981
ACNPN	ND	ND	129	ND	ND	ND	126
FLURN	ND	ND	117	ND	ND	ND	65
PHEN	8	5.5	1.5	0.02	1.5	3	3.3±1.3
ANTHR	ND	31.5	17.5	3.5	10.8	11.5	15±4.7
FLRTH	ND	10.5	16.5	2	3	4.5	7.3±2.7
PYR	17.5	10.3	34.5	13	17	24	19±3.5
CHRY	1.5	2	1.5	ND	2.5	8.5	3.2±1.3
BbF	11.5	26	27	6	18.5	10	16.5±3.5
BkF	15.5	10.5	2.5	10.5	1.5	6	7.8±2.1
BaP	3	64	6	3	2.5	ND	15.7±12
DahA	5.5	77.5	5.5	ND	11	1.6	15.7±12
BghiP	ND	128	ND	3.1	ND	4.1	20.2±14.3
$I_{123cd}P$	ND	ND	ND	ND	ND	ND	72±41
Total	127	6,351.3	3,952.5	347.6	119.8	640.7	1,923±1,069

*SE : Standard Error, , ND: Not detected

vivo. More knowledge of biochemical and microbial processes of PAHs in sediment and mussels will be needed to fully understand the fate of PAHs.

PAHs in both sediment and organisms can be easily affected by weather condition, and they are easily photochemically oxidized and removed from the body tissue of mussels via excretion, evaporation, physical weathering, etc. (Lee *et al.*, 1978; Barth, T. 1984. Weathering of crude oil in natural marine environments: The concentration of polar degradation products in water under oil as measured in several field studies. *Chemosphere.*, 13 : 67~86. Tjessem and Palmork, 1984; Berthou *et al.*, 1985; Ducreux *et al.*, 1986)

Total PAHs

Total PAHs in sediment ranged from 136.5 to 5,607.5 ppb (mean 1,234ppb) and those in mussels ranged from 119.8 to 3,952ppb (mean 1,923ppb) in the study area. A comparison of the total PAHs in sediment versus mussels is given in Fig. 5. Despite the variability of total PAHs in sediment and mussels in 6 sites, the

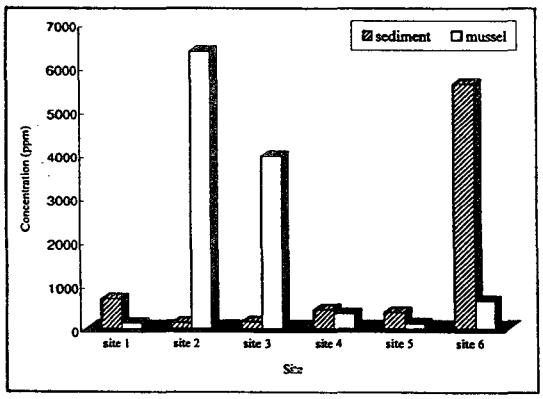

Fig. 5. Total concentration of PAHs in sediment versus mussels

PAHs in sediment from 4 sites (site 1, 4, 5 and 6) were 1.2~9 times higher than those in mussels. The levels of PAHs in marine ecosystem are well documented in world over, however, there are so limited amount of data regarding PAHs in organisms in Korea. The total PAHs in organisms observed in Chinhae bay, Korea (Lee, 1997), ranged from 498 to 2,060ppb wet wt (mean 760ppb), which is generally very similar to the results of the present study. Kahng (1995) reported the PAHs in sediment ranging from 145 to 3,362 ppb in Chinhae bay, which also shows similar levels of PAHs to the present study.

Comparison of PAH concentrations with other researches world over

The total PAH content in mussel in the Gulf of Naples from Italian Central Mediterranean coasts (Cocchieri *et al.*, 1990) varied from 2 to 60 ppb. Most PAH in the Gulf of Naples (Table 5) were very similar to those in the Southeast Sea, Korea, except NPTHL and ANCPL. NPTHL was not detected in the Gulf of Naples, whereas I123cdP was not detected in the Southeast Sea, Korea. ANCPL in the Southeast Sea, Korea was much higher than that in Naples.

Among the PAH contents analyzed in Yaquina Bay, the concentrations of PHEN, FLRTH and PYR were very higher than those in the Southeast Sea, Korea, while BbF and BghiP in the present study were a little higher than those in Yaquina Bay. And the concentration of NPTHL, ANCPL, ANCPN and FLURN in Lake Michigan were lower than those in the present study but the rest of the PAHs in Lake Michigan were higher than those in the Southeast Sea, Korea. In addition, PAH concentrations in mussel and surface sediment from Saudfjord were generally higher than those in the southeast Sea, Korea (Table 5).

In summary, it can be said that the levels of PAHs in mussels and sediment in the Southeast Sea, Korea are relatively low.

The sources of PAHs

Pyrogenic or combustion-derived PAH assemblages are relatively enriched in three- to five ring PAH compounds, whereas uncombusted fossil fuels are highly enriched in the two- to three ring PAHs (Boehm and Farrington, 1984). As shown in table 3 and 4, most PAH of 2~3 ring were higher in concentration than those of 3~5 ring (Fig. 7).

The ratio of naphthalene to phenanthrene is particularly diagnostic for inputs of fresh petroleum. While phenanthrene compound may be of petrogenic, or diagenetic in origin, naphthalene compound are characteristic of fresh crude oil. The N/P (Naphthalene/ Phenanthrene) ratio is much greater than 1.0 for most petroleums and decreases up to below 0.2 in clean sediments (Steinhauer and Boehm, 1992). These ratios are useful in defining the hydrocarbon composition of the marine organisms and sediments, and in distinguishing the relative importance of petroleum-derived (petrogenic) hydrocarbons versus biologically derived (biogenic) or combustion-derived (pyrogenic) hydrocarbons.

In this study, the N/P ratio ranged from 8.25 to 123 in sediment, and in mussels, it ranged from 0.8 to 825 (Fig. 6). We would suggest that the PAHs in mussels and sediment in the study area is mostly petroleum-derived. Also the relative abundance of two- to three- ring PAHs

Table 5. Comparison of PAHs reported from other researches world over

Sample and location	Concentration(μg/kg, Mussel: we wt, Sediment: dry wt)														
	NPTHL	ANCPL	ANCPL	FLURN	PHEN	ANTHR	FLRTH	PYR	CHRY	BbF	BbF	BaP	DahA	BghiP	I123cdP
Mussel from Present Sudy (Mean Concentration)	81	1068	126	65	3.3	15	7.3	19	3.2	16.5	7.8	15.7	20.2	72	ND
Mussel from Gulf of Naples[1]	ND	60	36	5	4	5	21	24	13	46	4	5	20	22	2
Mussel from Yaquina Bay[2]	NA	NA	NA	NA	1290	ND	260	135	ND	2	ND	38	ND	26	ND
Mussel from Saudfjord (wer wt)[3]	23	7	NA	413	NA	NA	NA	NA	3350	NA	1821	663	82	228	NA
Sediment from Present study (Mean Concentration)	226	149	1090	75	4.8	19	44	45	1.3	1.9	11.7	2.3	12	5.5	1.8
Sediment from Saudfjord[4]	483.8	93.8	31.8	36.0	2175.2	729.7	2134.9	1517.1	4161.2	2540.2	6312.9	1969.6	8703.0	NA	NA
Sediment from Lake Michigan[5]	10.2	29.1	21.27	7.1	209.6	36.79	495.90	399.87	347.86	614.52*		268.11	370.78	302.39	49.99

ND: not detected, NA: not analyzed. *BkF+BaF.

[1]Cocchieri et al., 1990 [2]Mix and Schaffer, 1983 [3,4])Bjø rseth et al., 1979 [5]Simcik et al., 1996

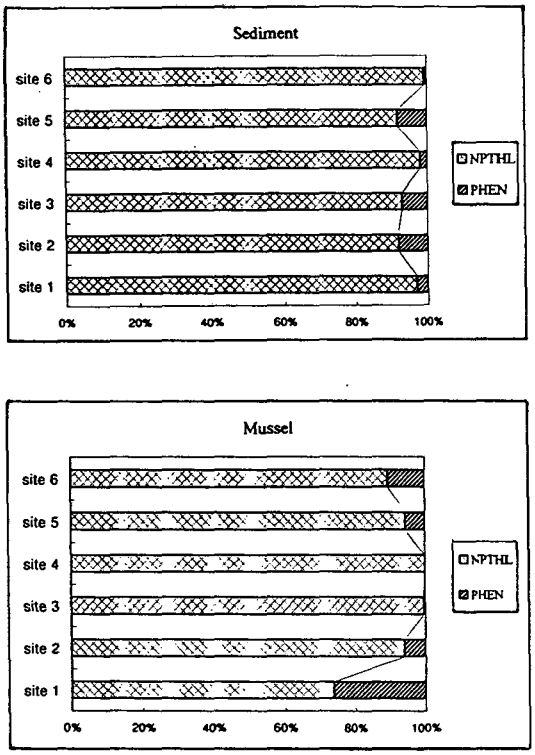

Fig. 6. NPTHL/PHEN(N/P) ratio of each site in the study area for the sediment and mussels

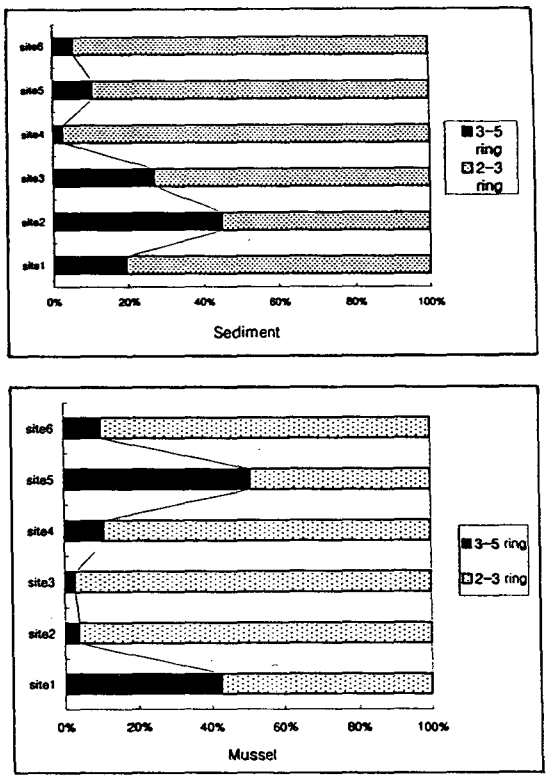

Fig. 7. The relative abundance of 2-3 ring vs. 3-5 PAHs ring in sediment and mussels in the study area

in sediment and mussels ranged from 58 % to 90 % of the total (Fig. 7). This confirms the idea that the major source of PAHs in the study area is not combustion-derived. Rather, the major source of PAHs in the study area could be domestic and industrial effluents containing uncombusted fossil fuels, and engine or fuel oil spilled from the vessels navigating.

Seasonal variations of PAH concentrations in sediment, mussels, seawater as well as other marine organisms in the study area need to be investigated in the future in order to support the above conclusion.

CONCLUSION

NPTHL and ANCPL were dominant PAH compounds in sediment and mussels in the Southeast Sea, Korea.

The concentration of PAH analyzed by HPLC with uv/vis detection in sediment ranged from 1 to 4,125 ppb (mean 112ppb), and in mussel, they ranged from 1.5 to 5,900 ppb (mean 108ppb) in the study area. Compared with other researches world over, the PAH levels in mussels and sediment of the study area is thought to be relatively low.

The concentration of high molecular weight

PAHs (3~5 ring) in mussels are higher than those in sediment, whereas low molecular weight PAHs (2~3 ring) in sediment are a little higher than those in mussel. It may be presumed that the higher the molecular weight of PAHs are, the easier thay can be attacked by microorganisms in sediment than they are *in vivo*. However, we need more valuable and sufficient data to support this.

According to N/P (Naphthalene/Phenanthrene) ratio in mussels and sediment of the study area, and to the ratio of 2~3 ring to 3~5 ring, we expect that the major sources of PAHs in this study area is rather petroleum-derived.

ACKNOWLEDGEMENT

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