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Polycyclic Aromatic Hydrocarbons (PAHs) in Seawater and Marine Sediments from Mokpo Coast in Korea

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Polycyclic aromatic hydrocarbons (PAHs) were determined in seawater and sediment from Mokpo coastal waters of Korea. Concentrations of PAHs in seawater and sediments ranged from 15.9 to 49.5 ng/L and from 4.79 to 511 ng/g dry weight, respectively. PAH concentrations measured in our study were lower than those in other locations in Korea and other countries. The PAH contamination was associated with intensive shipping and industrial activities. Based on diagnostic ratios and multivariate statistical analysis, the major source of PAHs in marine sediments was combustion processes, with a minor petrogenic input. There was no significant correlation between total organic carbon (TOC) and PAHs; however, there was high correlation between PCDD/Fs and PAHs, suggesting that these chemicals have similar sources and movements in the surveyed areas.

Key words : PAHs, Mokpo, seawater, sediment, total organic carbon (TOC)

1. Introduction

Mokpo, located on the southwest corner of Korea, has Muan in the north and a number of islands in the west coast. Mokpo coast is characterized by muddy flat with wide ranges of tide. Fresh water flows into this coast mainly from the Yeongsan River at the eastern end of the coast. The Yeongsan, Yeongam and Geumho embankments has been constructed for drainage since 1981.[1)] The inner part of the coast has the Daebul industrial complex, which was founded in 1996 and produces chemical products, nonferrous metals, machinery, and food products.[2)] Samho industrial complex located on the outer part of the coast comprised of shipyards and machine factories. Hence, the rapid industrialization of this area can be accompanied by a significant environmental deterioration that has led to a variety of social and health problems.

Polycyclic aromatic hydrocarbons (PAHs) are ubiquitous environmental pollutants of public concern and are derived mainly from anthropogenic activities.[3)] PAHs are generated by incomplete fuel combustion,[4)] domestic and industrial wastewaters,[5)] and spillage of crude oil and its refined products.[6,7)] Because of their carcinogenic and mutagenic potential,[8)] PAHs have been extensively studied in various environmental and biological compartments.[9,10)]

PAHs derived from various sources can be transported to aquatic environments via riverine inputs and atmospheric deposition.[11,12)] Concentrations of dissolved PAHs in seawater are very low, because they have hydrophobic character based on high octanol-water and organic carbon adsorption partition coefficients.[13)] PAHs in marine coastal waters adsorb strongly to suspended particles and deposited to bottom sediments. Consequently, analysis of sediment can provide information on

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recent sources of these contaminants in the marine environment. In addition, investigation on seawater contamination is necessary to grasp present pollution and to design future strategy for environmental management from the risks of toxic PAHs. Until now, the emphasis on environmental contamination for Mokpo coastal waters has only been focused on the water quality[2] and trace metals.[14] To our knowledge, the present study is a first effort to investigate concentrations and potential sources of PAHs in Mokpo coastal waters. This study was conducted as part of a nationwide survey of persistent organic pollutants (POPs) in Korea's coastal environment. The objective of this study was to describe the concentrations and sources of PAHs in seawaters and marine sediments from Mokpo coast of Korea.

2. Materials and Methods

2.1. Sampling strategy

Surface sediments (04 cm) were sampled at 29 locations from Mokpo coast in March 2006 (Fig. 1). Surface seawaters were sampled at 21 locations from Mokpo coast, excluding the outer part of the Mokpo

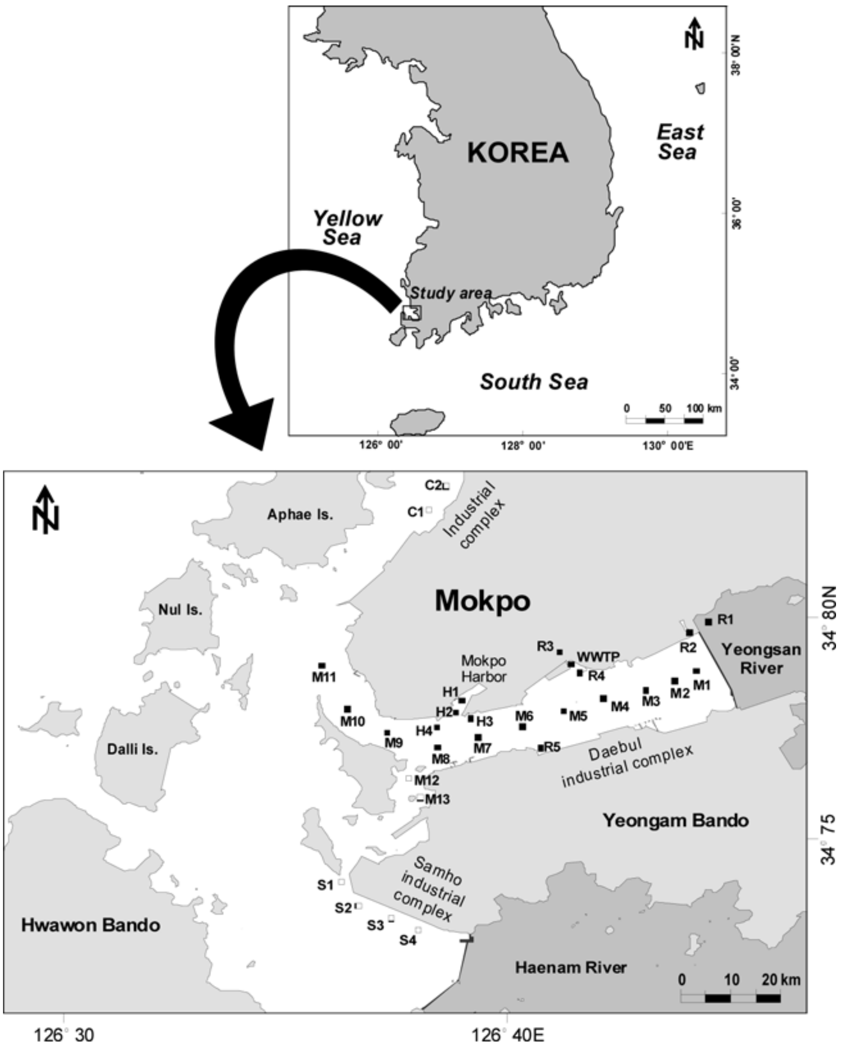

Fig. 1. Map showing sampling locations (enlarged map) of seawaters and marine sediments from Mokpo coastal waters in Korea. Black box (■) represents the locations collected both sample matrices and white box (□) represents the locations collected only sediment samples.

coast. These samples were collected systematically from the inner to the outer parts of the coast (Stations M1M13), harbor zone (Stations H1H4), Yeongsan River (Station R1), a location close to a wastewater treatment plant (Station WWTP) and some streams (Station R2R5) into the Mokpo coast. Our sampling area covered the locations near Samho industrial complex (Stations S1S4) and small industrial complex (Stations C1C2), which are located in the northern part of the Mokpo City. Surface seawaters were collected using a bucket sampler and transferred to polyethylene bottles of 10 L in a refrigerator. Surface sediments were sampled by using a box-corer deployed from a research vessel. The collected sediment samples were wrapped individually in aluminum foil and then frozen immediately in a refrigerator on board the vessel. Both environmental samples were transported to the laboratory and were kept in a freezer at 20°C until further analysis.

2.2. Sample preparation

Detailed descriptions of seawater and sediment samples extraction and clean-up procedures have been reported elsewhere.[12,15] Briefly, seawater samples of 4 L were extracted twice by liquid-liquid extraction using a mixture of methylene chloride (ultra residue analysis, J. T. Baker, Phillipsburg, NJ, USA) and hexane (ultra residue analysis, J. T. Baker) (1:1 100 mL). Marine sediments were extracted in a Soxhlet apparatus using 200 mL of 10% acetone (ultra residue analysis, J. T. Baker) in toluene (ultra residue analysis, J. T. Baker) for 24 h after spiking with 7 species of internal standards (d_8-naphthalene, d_8-acenaphthylene, d_{12}-fluoranthene, d_{10}-phenanthrene, d_{10}-pyrene, d_{12}-benzo[a]pyrene and d_{12}-benzo[g,h,i]perylene; ES-2044, Cambridge Isotope Laboratories, Andover, MA, USA). Both extracts were cleaned by passage through activated silica gel (neutral, 7734, 70-230 mesh, Merck, Darmstadt, Germany) column with successive eluants of hexane and 15% methylene chloride in hexane. The eluants were concentrated to approximately 1 mL and were evaporated at room temperature to 50 to 100 μL. The residues were dissolved with 50 μL *n*-nonane (pesticide analysis grade, Fluka, Switzerland) for instrumental analysis.

2.3. Instrumental analysis

Detailed descriptions of instrumental analysis have been described elsewhere.[12,15] Briefly, identification and quantification were performed by using a gas chromatography (Agilent 6890) interfaced with mass spectrometer detector (Agilent 5973N). The capillary column used was a DB5-MS (30 m length, 0.25 mm inner diameter, 0.25 μm film thickness; J&W Scientific). The mass spectrometer was operated under the selected ion monitoring mode using molecular ions of PAHs. In this study, 16 non-alkylated PAHs (48755-U; Supelco, Bellefonte, PA, USA) recommended as priority pollutants by the United States Environmental Protection Agency (US EPA) were analyzed. These include 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).

The total organic carbon (TOC) content in marine sediments was analyzed using a CHN Elemental Analyzer (PerkinElmer, Model 2400; Boston, MA, USA), after removal of calcium carbonate with 1 N HCl. Grain size analysis was performed using a particle size analyzer (Sympatec Windox4; Sympatec GmbH, Clausthal-Zellerfeld, Germany).

2.4. Quality assurance/quality control (QA/QC)

All of the deuterated internal standards were detected with no interferences. The recovery for seawater and sediment samples was 75 ± 17% and 82 ± 22%, respectively. Solvents injected before and after the injection of standards showed negligible contamination or carryover. Procedural blanks of seawater and sediments were processed in the same way as the samples. Blanks did not contain quantifiable amounts of the target compounds. The calculated detection limit (S/N = 3) for individual PAH was 1 ng/g dry weight.

3. Results and Discussion

3.1. Sediment characteristics

Grain size and TOC in marine sediments from Mokpo coastal waters are summarized in Table 1. Sediments collected in our study were fine clay to fine sand, with grain sizes of 5.1-17.6 μm. With exception of some locations such as Stations R1, R2 and M13, all of the sediment samples showed homogenous distribution of grain size, with values less than 10 μm. The TOC in marine sediments ranged from 0.4 to 2.4% with a mean value of 0.8%. Station R2 (Samhyang River; 2.4%) had the highest content of TOC in our study, however other locations contained low levels of TOC with values less than 1.0%. Although PAHs are expected to be associated with organic-rich particles, the correlation between the concentrations of PAHs and TOC in marine sediments was not high ($r = 0.363, p = 0.053$). This seems to be due to the combined effect of local contaminant sources, transport, mixing and deposition. Koh *et al.*[16] reported no significant correlation ($r = 0.58, p = 0.079$) between PAHs and TOC in coastal sediments from Yeongil Bay. However, a moderate correlation ($r = 0.597, p = 0.001$) between PAHs and PCDD/Fs were found in the sediments from Mokpo coastal waters, indicating that these contaminants are derived from the similar sources in the surveyed areas.

3.2. PAHs in seawater and sediments

Concentrations of PAHs in seawater and marine sediments from Mokpo coastal waters are summarized in Table 1. All of the PAH compounds were detected in all the seawater and sediment samples taken from this study. The highest concentrations of PAHs were found at Stations M7, M4, R4, R1 and M11, while the lowest concentrations of PAHs were found at Stations M5, M8 and H4 (Fig. 3). However, in general, there was no great difference in PAH contamination among the sampling locations in Mokpo coast. The total concentrations of PAHs (the sum of 16 PAH compounds) in seawater from Mokpo coast ranged from 15.9 to 49.5 ng/L with an average of 28.6 ng/L. The sum of the six potential carcinogenic PAHs (the sum of BaA, BbF, BkF, BaP, InP, and DbA), as proposed by the International Agency for Research on Cancer (IARC),[8] accounted for 40% of the

Table 1. Summary of PAH concentrations in seawater and marine sediments and sample characteristics from Mokpo coast in Korea

	Min	Max	Median	Mean±SD
Seawater (ng/L)				
NaP	1.99	7.26	3.91	3.93 ± 1.30
AcPy	0.36	9.43	3.69	4.11 ± 2.65
AcP	0.07	1.82	0.48	0.62 ± 0.45
Flu	0.61	1.89	1.16	1.16 ± 0.30
PhA	2.31	5.92	3.55	3.43 ± 0.85
AnT	0.17	1.38	0.44	0.50 ± 0.32
FluA	0.31	0.91	0.45	0.47 ± 0.16
Pyr	0.30	1.26	0.44	0.50 ± 0.22
BaA	0.11	2.52	0.96	0.96 ± 0.64
Chr	0.25	4.20	1.43	1.38 ± 0.95
BbF	0.11	24.7	2.59	5.47 ± 7.33
BkF	0.14	5.13	0.54	1.13 ± 1.27
BaP	0.20	4.48	0.38	0.67 ± 0.93
InP	0.08	5.66	0.93	1.14 ± 1.30
DbA	0.13	11.3	1.71	1.92 ± 2.41
BghiP	0.08	6.41	0.75	1.19 ± 1.60
ΣCPAH	0.87	30.4	8.35	11.3 ± 9.64
ΣPAH	15.9	49.5	24.9	28.6 ± 10.8
Sediment (ng/g dry weight)				
NaP	0.11	17.8	1.71	2.50 ± 3.37
AcPy	0.01	0.53	0.10	0.16 ± 0.14
AcP	0.00	1.80	0.05	0.17 ± 0.36
Flu	0.08	11.1	1.44	2.20 ± 2.66
PhA	0.43	70.6	9.03	12.8 ± 15.3
AnT	0.08	18.9	1.61	2.31 ± 3.55
FluA	0.94	68.7	14.0	19.0 ± 17.4
Pyr	0.60	82.0	13.3	16.0 ± 18.1
BaA	0.20	32.5	4.50	6.11 ± 7.04
Chr	0.46	49.8	9.01	12.2 ± 11.7
BbF	0.82	61.2	15.5	17.8 ± 14.9
BkF	0.21	21.6	3.91	5.26 ± 4.92
BaP	0.10	32.1	2.59	5.14 ± 7.54
InP	0.33	23.9	5.35	7.74 ± 6.52
DbA	0.06	4.77	0.88	1.30 ± 1.25
BghiP	0.32	25.8	5.74	7.30 ± 6.09
ΣCPAH[a]	1.76	176	33.6	43.3 ± 40.7
ΣPAH[b]	4.79	511	95.9	118 ± 113
Sediment characteristics				
Grain size (μm)	5.14	17.6	6.04	7.32 ± 3.19
TOC (%)	0.42	2.42	0.69	0.76 ± 0.35

[a]The sum of the six carcinogenic PAHs of IARC recommendation.
[b]The sum of total sixteen PAHs.

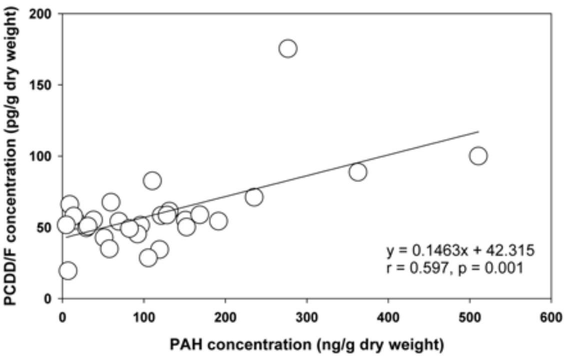

Fig. 2. Correlation between the concentrations of PAHs and PCDD/Fs in marine sediments from Mokpo coastal waters in Korea.

total concentrations of PAHs in seawater. The total PAH concentrations in our study were comparable to those from Rhone Delta (average 12 ng/L)[17] and San Francisco Bay (average 25 ng/L).[18] The PAH concentrations in coastal water from other studies such as Tamar estuary (83 ng/L),[19] Yangtze estuary (183 ng/L),[20] and Seine River (204 ng/L)[21] were higher than those in our study.

The total PAH concentrations in the sediments from Mokpo coastal waters ranged from 4.79 to 511 ng/g dry weight with an average of 118 ng/g dry weight (Table 1). Concentrations of carcinogenic PAHs were 43.3 ± 40.7 (average ± SD) ng/g dry weight, accounting for 37% of the total PAH concentrations in the sediments. The PAH concentrations measured in our study were lower than those reported at other locations from several Korean coasts. Concentrations of PAHs in marine sediments from some industrialized bays in Korea (38-2413 ng/g dry weight),[15] Ulsan Bay (6.0-2396 ng/g dry weight),[22] Yeongil Bay (205–10,686 ng/g dry weight),[23] Masan Bay (207-2670 ng/g dry weight),[24] and Gyeonggi Bay (9.1-1400 ng/g dry weight)[25] showed higher levels than those in Mokpo coastal waters. In addition, these PAH concentrations in Mokpo coastal sediments did not exceed the sediment quality guidelines such as effect range-low (ERL; 4000 ng/g dry weight) and effect range-medium (ERM; 45000 ng/g dry weight) proposed by National Oceanic and Atmospheric Administration (NOAA).[26] Therefore, the sediment contamination by PAHs in Mokpo coast seemed to be low and safe in comparison to that in other aquatic environment of Korea.

The PAH contamination distributions in the sediments from Mokpo coastal waters did not show a clear decreasing trends from inner to outer locations (Fig. 3). The highest concentration of PAHs was found at harbor zones such as Stations H1 and H2, suggesting that harbor and shipyard activities contributes to the PAH contamination in this coast. Station R2 (Samhyang River) had a relatively high concentration of PAHs, indicating that this river can be the main route for PAH contamination in Mokpo coastal waters. However, the sediment from Yeongsan River (Station R1) had a relatively low concentration (51.3 ng/g dry weight),

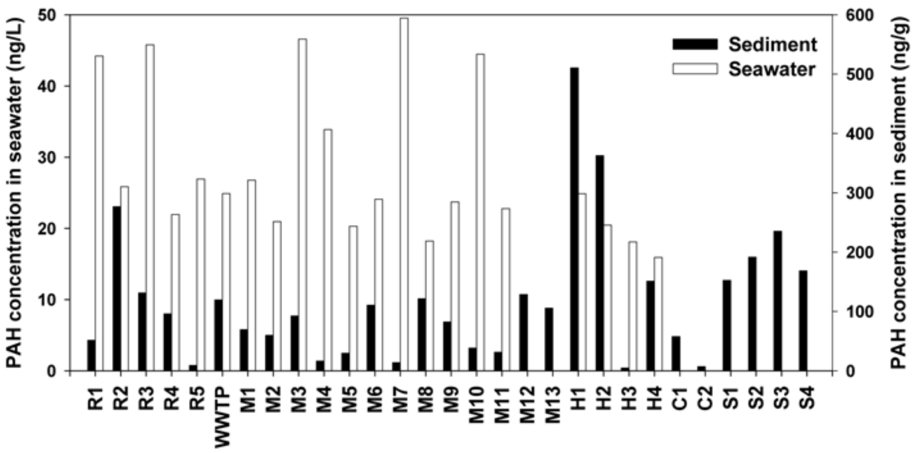

Fig. 3. Distribution of PAH concentrations in seawater and marine sediments from Mokpo coast in Korea. Detailed sampling locations are described in Fig. 1.

suggesting that this river is not the main source of PAH contamination in Mokpo coast. The sediments (Stations S1–S4) collected near Samho industrial complex showed relatively high concentrations (152-235 ng/g dry weight). The PAH concentration in the sediment from near a WWTP was relatively moderate (119 ng/g dry weight). Consequently, the main source of PAHs in Mokpo coastal waters is intensive shipping and industrial activities.

3.3. PAH profiles in seawater and sediment

The PAH profiles in seawater and marine sediment from Mokpo coast are shown in Fig. 4. Seawater samples were dominated by the lower-molecular-weight PAHs such as two and three ring aromatics compared to those of sediments, while the higher-molecular-weight PAHs were dominant compounds in the sediments. In the seawater, the predominant compounds were AcPy, NaP, BbF and PhA, collectively accounting for 60 ± 38% (average ± standard deviation) of the total PAH concentrations. In the sediments, FluA, BbF, Pyr, Chr and PhA collectively accounted for 67 ± 13% (average ± standard deviation) of the total PAHs. These PAH profiles are similar to those reported for seawater and marine sediments from several Korean coasts.[15,23] The difference of PAH profiles between both environmental matrices may be due to the effect of degradation of PAHs in aquatic environment. In general, the persistence of PAHs in the environment increases with increasing molecular weight. Approximately 80% of lower-molecular-weight PAHs can be lost by volatilization from surface seawater into the atmosphere.[27] Moreover, microbial degradation and biotransformation seems to be major environmental fate processes governing the accumulation profiles of PAHs in the aquatic ecosystem.[28,29]

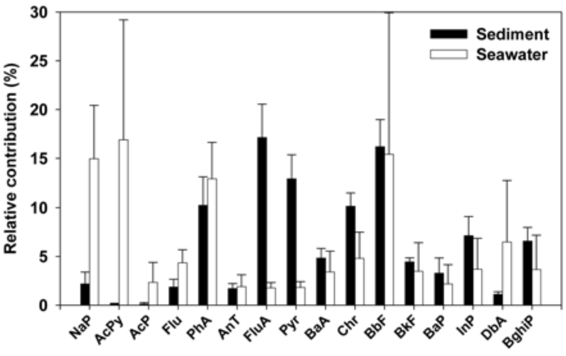

Fig. 4. Average normalized profiles of PAHs in seawater and marine sediment from Mokpo coast in Korea. Vertical lines represent standard deviations.

3.4. Sources of PAHs in Mokpo coastal waters

Although several constraints exist, molecular ratios of some marker PAHs have been used to identify sources of PAHs in marine environment.[30] The result of common diagnostic ratios (PhA:AnT and FluA:Pyr) investigated in this study is presented in Fig. 5. Due to their different physico-chemical characteristics, these diagnostic ratios give useful information on the PAH sources.[31] The ratio of PhA:AnT < 10 and FluA:Pyr > 1 implies pyrolytic origin, and the ratio of PhA:AnT > 10 and FluA:Pyr < 1 indicates petrogenic origin of PAHs.[32] Most of the sediment samples had the values of PhA:AnT < 10 and FluA:Pyr > 1, indicating that combustion process of fossil fuels is the major source of PAHs in Mokpo coastal waters. However, some locations such as Stations H1, M7, R2, R3 and WWTP showed values of FluA:Pyr < 1 and/or high values of PhA:AnT. This suggests that both pyrolytic and petro-

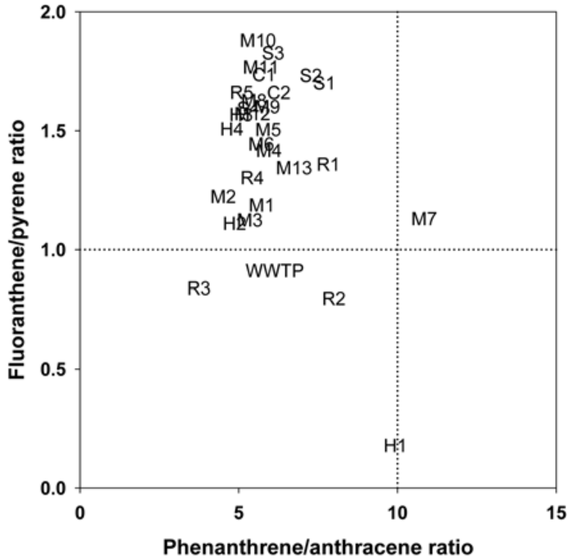

Fig. 5. Relationship between phenanthrene/anthracene and fluroranthene/pyrene in marine sediments from Mokpo coastal waters in Korea.

genic input as sources in Mokpo coast and the predominance of pyrolytic origin in these locations.

To characterize the spatial variability of PAH profiles in marine sediments from Mokpo coastal waters, we performed two-dimensional non-parametric multidimensional scaling (MDS) ordination using PRIMER for Windows (PRIMER Version 5.2.9, Plymouth, UK). The 16 PAH compounds measured in the sediments were subjected to MDS ordination, using the Bray-Curtis similarities calculated from square-root transformed data (Fig. 6). This multivariate statistical technique has been used to determine the spatial variability of chemical compositions in sediments.[33,34] The plot had a stress of 0.01, with values less than 0.1 representing a good ordination with little chance for misinterpretation.[35]

Four clusters were identified on the variable plot, each containing data grouped according to their profiles of PAHs (Fig. 7). Although it is difficult to identify a clear difference of PAH profiles among groups by MDS ordination, most sampling locations were located close inner or near the circles except some locations. This suggests that near sampling locations in Mokpo coast have similar sources of PAHs. The first cluster (Group A) included the sediments from the rivers (Stations R1–R4) and a WWTP. The PAH profiles were dominated by the lower-molecular-weight aromatics such as NaP, Flu, PhA and Pyr, compared to the profiles of other clusters.

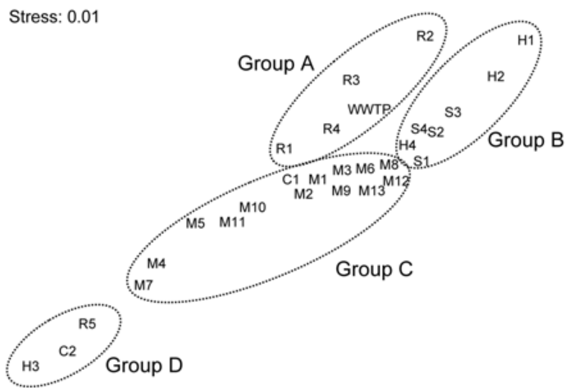

Fig. 6. Non-parametric multidimensional scaling (MDS) ordination plots of PAH compound distribution in marine sediments from Mokpo coastal waters in Korea.

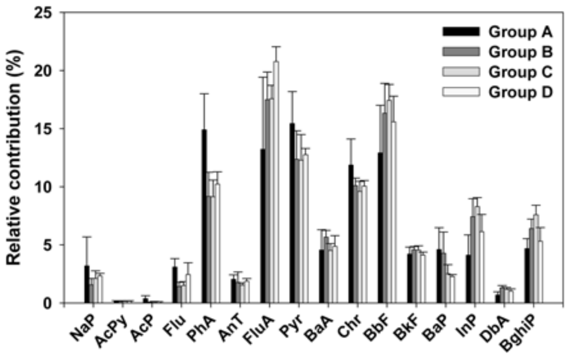

Fig. 7. Comparison of PAH profiles for each group clustered by MDS ordination results. Results for each compound were normalized to the total concentration of PAHs in the sediments. Vertical lines represent standard deviations.

Lee et al.[11] reported that the lower-molecular-weight PAHs mainly are derived from riverine discharge into the coastal waters, while the higher-molecular-weight PAHs primarily are transported by atmospheric deposition. The second group, Group B, was clustered by the sediments from the harbor (Stations H1, H2, H4) and Samho industrial complexes (Stations S1–S4), which comprised of shipyards and machine factories. The PAH source of these locations seems to be associated with shipping and shipyard activities. However, these PAH profiles are similar to those of third cluster (Group C), which represented Stations M1–M13 and C1. The PAH profiles in the sediments were characterized by the predominance of the higher-molecular-weight aromatics, compared to those of other clusters, suggesting that atmospheric deposition is the main route for PAH contamination for these locations. [11] The fourth group (Group D) was clustered by some locations such as Stations H3, C2, and R5. Although these locations have different potential sources of PAHs, their PAH profiles were relatively similar with high FluA concentration. This suggests that there exists a specific source associated with these contaminants.

4. Conclusions

PAHs are widely distributed in Mokpo coastal waters of Korea. Concentrations of PAHs in seawater and marine sediments were lower than those in other locations in Korea and other countries. The PAH contamination was associated with intensive shipping and industrial activities. Based on diagnostic ratios and non-parametric MDS, a major source of PAHs in marine sediments was combustion processes, with a minor petrogenic input.

Acknowledgement

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