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ARTICLE

Occurrence, Distribution, and Risk Assessment of Polycyclic Aromatic Hydrocarbons in the Surface Water of the Dongjin River, Republic of Korea

Jae Kwon Son · Byung Yeoup Chung · Jeongryeol Jang · Jae Young Cho

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Abstract The occurrence, distribution, and ecological risk assessment of 15 polycyclic aromatic hydrocarbons (PAHs) were investigated in the Dongjin River water system from December 2010 to October 2012. Among the detected PAHs, the mean concentration of acenaphthylene was the highest. Other PAHs were detected at very low concentrations. The detection frequencies and concentrations of the 15 PAHs were generally higher in the winter season, indicating low water flow conditions and low temperature. The results of a survey of the origin of the PAHs using the Phe/Ant ratio and Fla/Pyr ratio clearly indicated a pyrogenic source. The risk quotient (RQ) values for the 15 PAHs in the Dongjin River water system were below 0.01–0.1, indicating little risk to the relevant sensitive aquatic organisms, including green algae and daphnids, by the target compounds. In particular, the RQ values of most of PAHs exceeded 0.1 for fish in all of the seasons at most of the sampling sites, which indicated that the fish were exposed to medium risk.

Keywords Dongjin River · ecological risk assessment · polycyclic aromatic hydrocarbons · Saemangeum

J. K. Son and B. Y. Chung contributed equally.

J. K. Son
Department of Agricultural Engineering, Chonbuk National University, Jeonju 561-756, Republic of Korea

B. Y. Chung
Advanced Radiation Technology Institute (ARTI), Korea Atomic Energy Research Institute (KAERI), Jeongeup 580-185, Republic of Korea

J. Jang
Rural Research Institute, Korea Rural Community Corporation, Ansan, 426-908, Republic of Korea

J. Y. Cho (✉)
Department of Bio-environmental Chemistry, Chonbuk National University, Jeonju 561-756, Republic of Korea
E-mail: soilcosmos@jbnu.ac.kr

Introduction

Polycyclic aromatic hydrocarbons (PAHs), also known as polyaromatic hydrocarbons or polynuclear aromatic hydrocarbons, are widespread organic pollutants present in the atmosphere, water, soil, sediment, sludge, and organisms (Guo et al., 2007; Lee et al., 2008; Ju et al., 2009; Qin et al., 2013). PAHs are persistent organic pollutants with very low solubility in water system (Kafilzadeh et al., 2011). PAHs are formed by the incomplete burning of hydrocarbons and are usually found as a mixture by adsorption onto airborne particles, which were then rebroadcast into the air as particles (Watabe et al., 2013). The sources of PAHs include automobile fuel exhaust, tire degradation, industrial emissions from catalytic cracking, air-blowing of asphalt, coal coking, domestic heating emissions from coal, oil, and wood, waste incineration, and biomass burning (Deng et al., 2006). Some PAHs are reported to be toxic, mutagenic, and carcinogenic materials (Lee et al., 2008; Qin et al., 2013).

Several researchers have studied the occurrence, distribution, and environmental risk assessment of PAHs in water systems. Among the Asian countries, researchers from China have conducted regular monitoring of PAHs in various river water samples (Guo et al., 2007; Sun et al., 2009; Guo et al., 2011). The distribution of PAHs in the aquatic systems of other countries, such as India (Malik et al., 2011), Iran (Kafilzadeh et al., 2011), Italy (Patrolecco et al., 2010; Montouri and Triassi, 2012), South Africa (Moja et al., 2013), Hungary (Nagy et al., 2012), Egypt (Nemr and Abd-Allah, 2003), and the USA (Zhang et al., 2007), have also been reported. At present, very limited investigations have been carried to evaluate the PAHs in the environment of Korea (Kim, 2000; Nam et al., 2002; Lee et al., 2005; Lee et al., 2008; Ju et al., 2009). Few studies have been carried out to monitor PAHs in the river water of Korea. During the past few decades, due to industrial development, urbanization, and growth in the population, the generation of sludge in Korea has increased from sewage treatment plants and waste water treatment plants (Ju et al., 2009). This sludge may contain toxic pollutants, including PAHs, which may reach the ocean through river or streams. Continuous monitoring of the presence of PAHs in the river water is important, because they have adverse effects on living organisms. Therefore, we conducted this study in order to investigate the levels of 15 PAHs, their patterns, distribution, ecotoxicological risk assessment, and relationship sources in water samples collected from the Dongjin River in Korea from December 2010 to October 2012.

Materials and Methods

Chemicals. The authentic PAH standards and surrogate compounds were purchased from Supelco (USA). The high-performance liquid chromatography-grade methylene chloride, methyl alcohol, acetone, and hexane were purchased from the Duksan Co. (Korea).

Study area. Dongjin River flows through the cities of Jeongeup-si and Gimje-si, Korea. The study area has a continental monsoon climate. The annual rainfall averages nearly 1,300 mm, most of which falls in late June through August. The winters are dry and cold, and there is low water flow. Dongjin River is 46 km in length and has a total watershed of 1,109 km^2, and the status of land use is upland (18%), paddy field (42%), forest (29%), land (7%), and other (4%). A detailed description of the study area, as well as the sampling locations under investigation, is provided in Fig. 1. Dongjin River has been used for irrigation water for paddy fields. The sampling sites Jeongeup [JE, N(35°63'95"), E(126°86'50")], Jooksan [JS, N(35°76'76"), E(126°81'11")], and Yeonji [YJ, N(35°56'84"), E(126°84'39")] were located upstream from the watershed, un-urbanization and agricultural areas; the Sangdong [SD, N(35°55'46"), E(126°86'92")] site was located approximately 3 km downstream from a large residential and agroindustry complex (plastic manufacturing, special vehicle manufacturing, small petrochemical complex, nonmetallic manufacturing, and textile industry); the Gongpyung [GP, N(35°58'37"), E(127°82'80")], Ongdong [OD, N(35°61'66"), E(126°97'86")], and Buryang [BR, N(35°71'85"), E(126°81'83")] sites were located midstream from the watershed; the Shintaein [ST, N(35°67'72"), E(126°88'92")], Taein [TI, N(35°64'89"), E(126°92'76")], and Baeksan [BS, N(35°70'55"), E(126°78'25")] sites were located only 4 to 6 km from the river mouth.

Sample collection and treatment. Water samples from 10 sites were collected bimonthly for 2 years from December 2010 to October 2012. Ten liters of water were collected from each sampling site. After shaking, the water samples were filtered through a 0.45-μm glass fiber filter using a filtration device consisting of a peristaltic pump (80EL005, Millipore Co., USA). Surrogate standards of acenaphthene-d_{10}, phenanthrene-d_{10}, chrysene-d_{12}, and perylene-d_{12} were spiked in the water samples to indicate the recovery before extraction. The water samples were extracted using a solid phase extraction (SPE) system. The C_{18} cartridges were prewashed with methylene chloride and conditioned with methanol and de-ionized water. A water sample of 4 L was passed through the SPE system and extracted. The cartridges were eluted with 20 mL of methylene chloride. The volume of the extracts was reduced using a vacuum rotary evaporator in a water bath and was adjusted to a volume of 1 mL with hexane. Internal

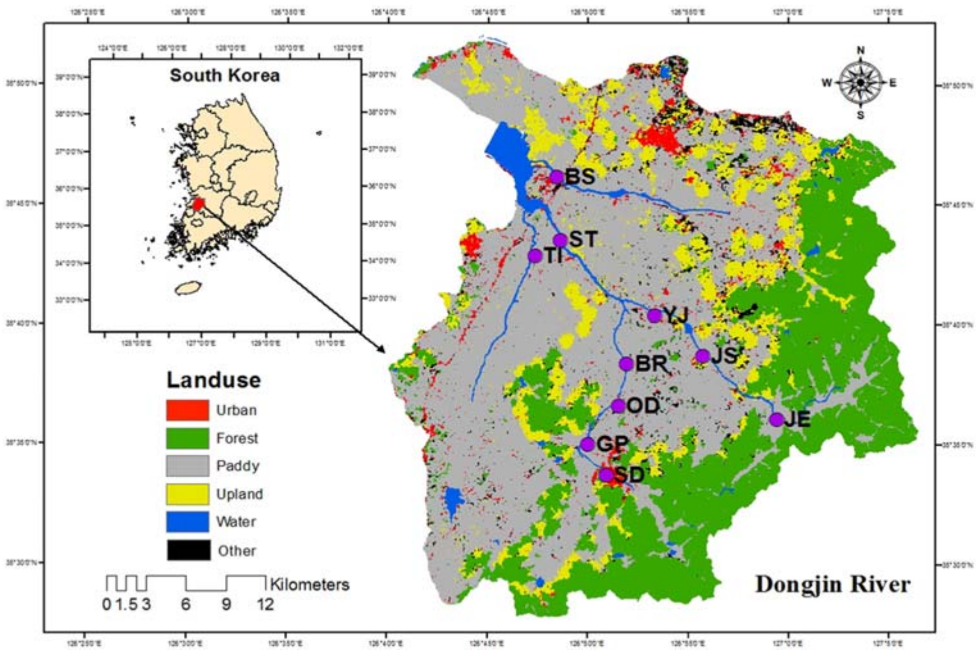

Fig. 1 Sampling sites along the Dongjin River water system, Korea.

standards dissolved in isooctane containing acenaphthene-d_8 and chrysene-d_{12} were added for the GC-MS analysis with a splitless injection.

Instrumental analysis and quality control. All samples were analyzed on a gas chromatograph with a mass spectrometer detector (Agilent 6890GC/5973MSD). A 30 m×0.25 mm i.d. with a 0.25-μm film thickness HP-5MS capillary column (Agilent Technology) was used. The column temperature was programmed to increase from 60 to 280°C at 5°C/min, and the temperature was kept isothermal for 20 min. MSD was operated in the electron impact mode at 70 eV and the ion source temperature was 230°C. The mass spectra were recorded using the selected ion monitoring mode. The 15 PAH compounds used in this study were: acenaphthene (Ace), acenaphthylene (Acy), fluorine (Flu), phenanthrene (Phe), anthracene (Ant), fluoranthene (Fla), pyrene (Pyr), benz(a)anthracene (BaA), chrysene (Chr), benzo(b)fluoranthene (BbF), benzo(k)fluoranthene (BkF), benzo(a)pyrene (BaP), dibenz(a,h) anthracene (DahA), indeno(1,2,3-cd) pyrene (IcdP), and benzo(g,h,i)perylene (BghiP). Quantification was performed by the internal standard method using a 15 PAH reference material mixture, with correlation coefficients for calibration curves that were all higher than 0.98. In the reference material mixture, the recoveries ranged from 80.5 to 102.1%, whereas the respective relative standard deviations ranged from 6.9 to 10.2%. The detection limits of this method ranged from 0.49 to 9.68 ng/mL in a sample on dry mass basis. All experiments were carried out in duplicate.

Ecological risk assessment (ERA). The potential risk of environmental pollutants can be estimated from an index of ERA. Generally, the risk quotient (RQ) is calculated from the ratio of a measured environmental concentration (MEC) and a predicted no-effect concentration (PNEC, ChV) using the lowest value for each endpoint, as shown in the following equation:

$$RQ = MEC/PNEC.$$

The PNEC value was obtained by dividing the lowest no observed-effect concentration (NOEC) for the most sensitive species by a safety factor, in which the default safety factors of 1,000 and 100 for the acute and chronic toxicity, respectively, were used to derive the PNECs (Zhang et al., 2012). In the present study, a safety factor of 100 was adapted in relation to the chronic toxicity of the PAHs as follows:

$$PNEC = NOEC/100.$$

The MEC data for the 15 PAHs was obtained from the measured concentrations, and the toxicity data was calculated from the ecological structure-activity relationships (ECOSAR) model (US EPA, 2011). The aquatic organisms from three different trophic levels: green algae, daphnids, and fish, were chosen as the ECOSAR model (Yan et al., 2013).

Results and Discussion

PAHs in the surface water of the Dongjin River. A total of 15 PAHs were analyzed at different sampling sites in the Dongjin River, Korea, during the period from December 2010 to October 2012. Table 1 summarizes the statistical data determined for the concentrations of the PAHs. Among the detected PAHs, the mean value of acenaphthylene had the highest concentrations, followed by acenaphthene, fluorene, and phenanthrene. The other PAHs were detected in very low concentrations. The highest PAH concentrations were located in the surface water from the SD district site, which is adjacent to Jeongeup agro-industry complexes and large residencial areas. In addition, the water flow was slow

Table 1 Summary of the 15 PAHs concentrations in the Dongjin River water system from December 2010 to October 2012 (n =120)

PAH compounds	Concentration (ng L^{-1})				
	Range	Mean	Median	Mode†	SD
Acenaphthene (Ace)	0-2.65	0.49	0.38	0	0.56
Fluorene (Flu)	0-2.69	0.67	0.46	0	0.77
Acenaphthylene (Acy)	0-3.14	0.68	0.34	0	0.82
Phenanthrene (Phe)	0-1.68	0.56	0.57	0	0.52
Anthracene (Ant)	0-1.96	0.43	0.37	0	0.45
Fluoranthene (Fla)	0-1.00	0.17	0.15	0	0.17
Pyrene (Pyr)	0-0.54	0.15	0.15	0	0.15
Benzo(a)anthracene (BaA)	0-0.11	0.03	0	0	0.03
Chrysene (Chr)	0-0.18	0.04	0	0	0.05
Benzo(b)fluoranthene (BbF)	0-0.06	0.01	0	0	0.02
Benzo(k)fluoranthene (BkF)	0-0.06	0.01	0	0	0.02
Benzo(a)pyrene (BaP)	0-0.07	0.01	0	0	0.02
Dibenzo[a,h]anthracene (DahA)	0	0	0	0	0
Indeno[1,2,3-cd]pyrene (IcdP)	0	0	0	0	0
Benzo[ghi]perylene (BghiP)	0	0	0	0	0

The mode (†) is the value that appears most often in a set of data.

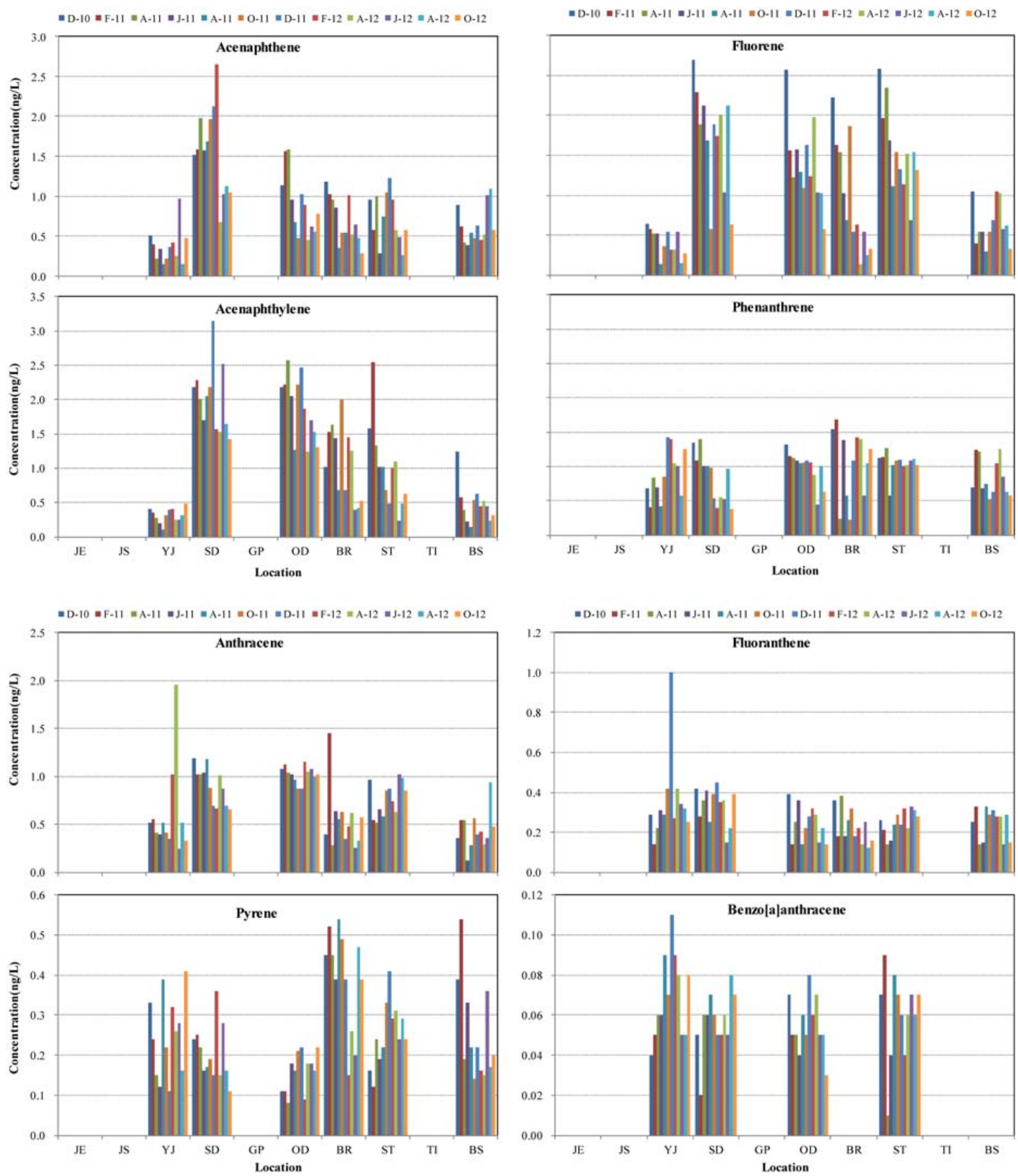

Fig. 2 Concentrations of PAHs in the Dongjin River water system from December 2010 to October 2012.

during December 2010 as it was the winter season. PAHs were not detected at the JE, YS, GP, and TI districts, which may be due to the presence of small residences, small agricultural lands, and a large forest. The deposition of PAHs in the surface water of the Dongjin River from December to February could be due to the increased atmospheric inputs in winter. Differences were observed in the total PAH concentrations between the urbanization and agro-industry complex districts (SD, OD, BR, and ST), and the un-urbanized and agro-forestry districts (JE, YS, GP, and TI). In our investigation, a low molecular weight 3-ring PAHs (Ace, Acy, Flu, Phe, and Ant) pattern predominated in the water samples, due to the relatively high vapor phase and water solubility (Sun et al., 2009). Intermediate molecular weight 4-ring PAHs (Fla, Pyr, BaA, and Chr) were partitioned between the vapor and particulate

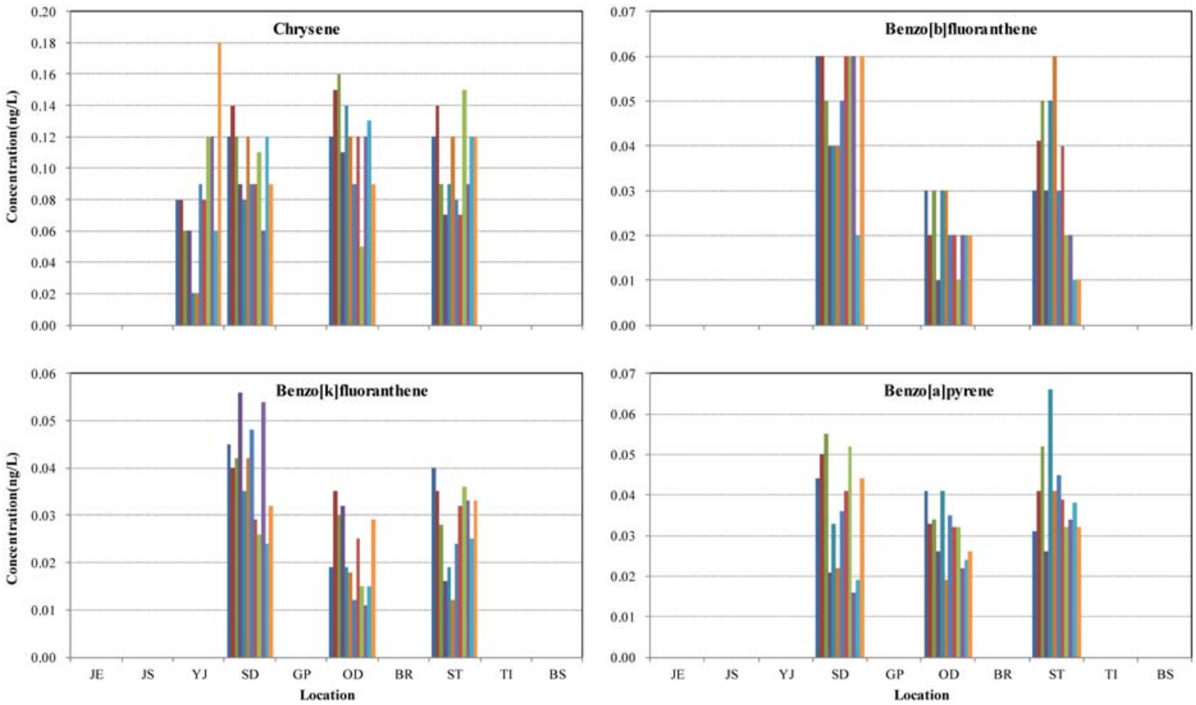

Fig. 2 Continued.

phases, depending on the atmospheric temperature (Srogi, 2007). The high molecular weight 5- to 6-ring PAHs (BbF, BkF, BaP, DahA, IcdP, and BghiP) were detected in low amounts at the SD, OD, and ST sites, and was not detected at other sites (Fig. 2). In addition to the atmospheric fallout due to the wet and dry deposition of particles, the PAHs enter surface waters especially via urban run-off, municipal effluents, industrial effluents, and oil spillage or leakage (Nagy et al., 2012). Atmospheric deposition and urban surface runoffs are important sources of PAHs (Zhu et al., 2004; Gigliotti et al., 2005; Deng et al., 2006; Sun et al., 2009). The detected concentrations of PAHs in the Dongjin River of Korea, which were lower than researchers have reported in other geographical regions, are listed in Table 2.

Sources of PAHs. PAHs primarily originate from anthropogenic activities and are usually formed through pyrogenic (fuel combustion) and petrogenic (crude oil discharge) sources (Montuori and Triassi, 2012). PAHs enter river environment systems primarily through deposition, urban runoff, municipal/industrial effluents, and oil leakage (Zhu et al., 2004; Gigliotti et al., 2005; Guo et al., 2007). In order to understand the fate of PAHs in the environment,

Table 2 Global comparison of PAH concentrations in surface water systems

Location	Year of sampling	n	Range (ng/L)	Reference
Dongjin River, Korea	2010-2012	15	0-53.9	This study
Danube River, Hungary	2007-2010	16	25-357	Nagy et al., 2012
Sarno River, Italy	2008	16	23.1-2,670.4	Montouri and Triassi, 2012
Gomti River, India	2004-2006	16	60-84,210	Malik et al., 2011
Tiber River, Italy	2007	6	23.9-72.0	Patrolecco et al., 2010
Pearl River, China	2002-2003	15	12.9-182.4	Luo et al., 2008
Mississippi River, USA	2004	16	62.9-144.7	Zhang et al., 2007
Tianjin River, China	2004	16	1,765-35,210	Cao et al., 2005
Hangzhou River, China	2002	10	989-9,663	Chen et al., 2004
Gao-ping River, Taiwan	1999-2000	16	10-9,400	Doong and Lin, 2004
Susquehanna River, USA	1997-1998	36	17-150	Ko and Baker, 2004
Alexandria Coast, Egypt	2002	7	103-523	Nemr and Abd-Allah, 2003
York River, USA	1998-1999	20	2.09-123	Countway et al., 2003
Gulf of Mexico, USA	1999	18	0.06-4,333.9	Mitra and Bianchi, 2003
German-Black Sea	2004	16	28-1,163	Nagy, 2007

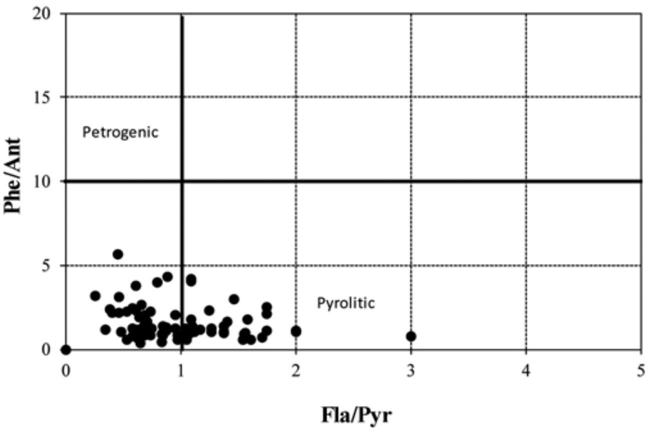

Fig. 3 Phe/Ant and Fla/Pyr ratio in the Dongjin River water system, Korea.

it is very important to identify the possible sources of PAHs in the Dongjin River system. In our study, the phenantherene/anthracene (Phe/Ant) and fluoranthene/pyrene (Fla/Pyr) ratios were calculated in order to identify the possible origin of the PAHs. The Phe/Ant concentration ratios have been widely used to identify combustion derived PAHs when pyrogenic PAHs usually have higher ratios of the less stable isomer i.e., anthracene, to the thermodynamically more stable one i.e., phenantherene (Zhang et al., 2007). A ratio of Phe/Ant <10 was usually regarded as an indication of pyrolytic sources, whereas Phe/Ant ratio >10 was primarily from petrogenic sources (Sun et al., 2009). Based on the average PAH data in the Dongjin River water system, the Phe/Ant ratio was calculated and it was below the value of 10 (Fig. 3). Therefore, it clearly indicated a pyrogenic source. The Fla/Pyr concentration ratio is also commonly used to distinguish between pyrogenic and petrogenic sources (Zhang et al., 2007). A ratio of Fla/Pyr <1.0 was usually attributed to petrogenic sources, while Fla/Pyr >1.0 suggested pyrolytic sources. In this study, the ratios of Fla/Pyr in the Dongjin River water system indicated that the PAHs were again primarily of pyrogenic origin. Therefore, traffic may be one of the principal reasons for the increased PAHs concentrations observed in the Dongjin River water system, Korea.

Ecotoxicological risk assessment. ERA is the key principle by which the risk of PAHs to the environment is understood. An ERA evaluates any potential harm that human activities have on living organisms within ecosystems (US EPA, 2001). In recent years, many researchers have focused on ERA to understand and predict the relationship between stressors and ecological effects in a way that is useful for environmental decision making. This assessment includes chemical, physical, and biological monitoring of water and sediment (Sany et al., 2012). In the early stages of risk assessment, the RQ, which is the quotient of the measured or estimated environmental concentration divided by the toxicant

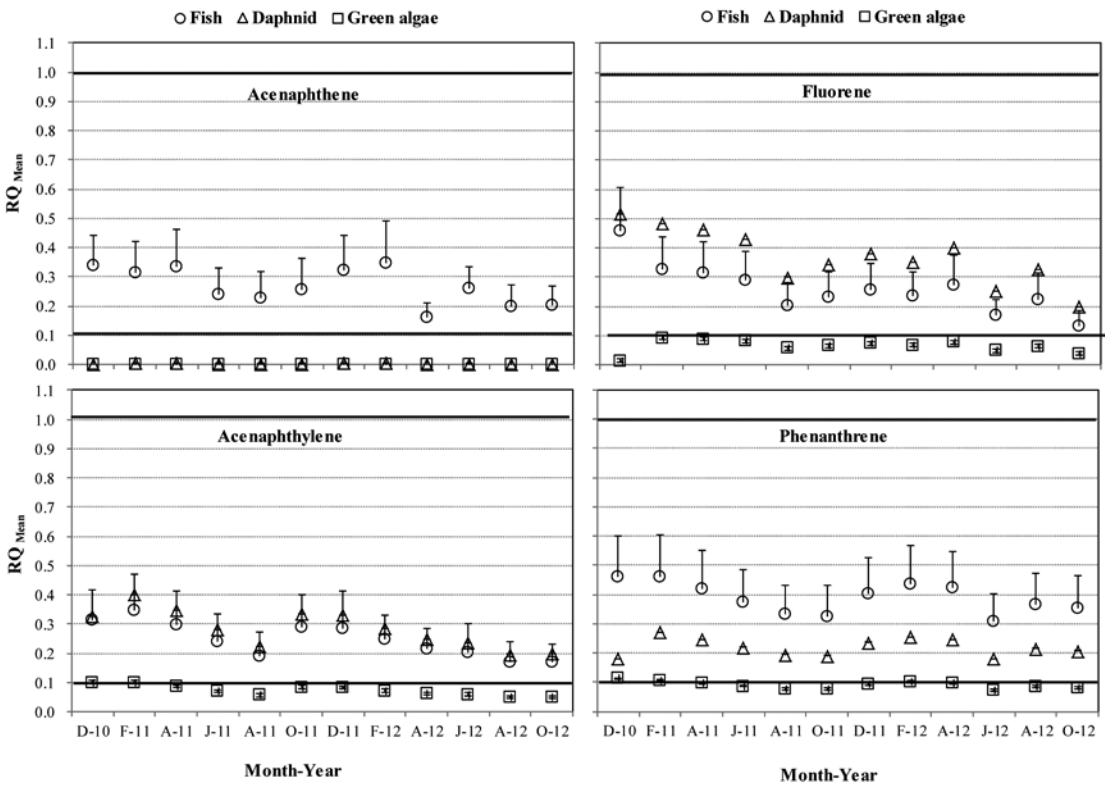

Fig. 4 Risk assessment of the PAHs in the Dongjin River water system from December 2010 to October 2012. The different symbols represent the median RQ values for the three different organisms, and the bars represent the maximum RQ values for those organisms.

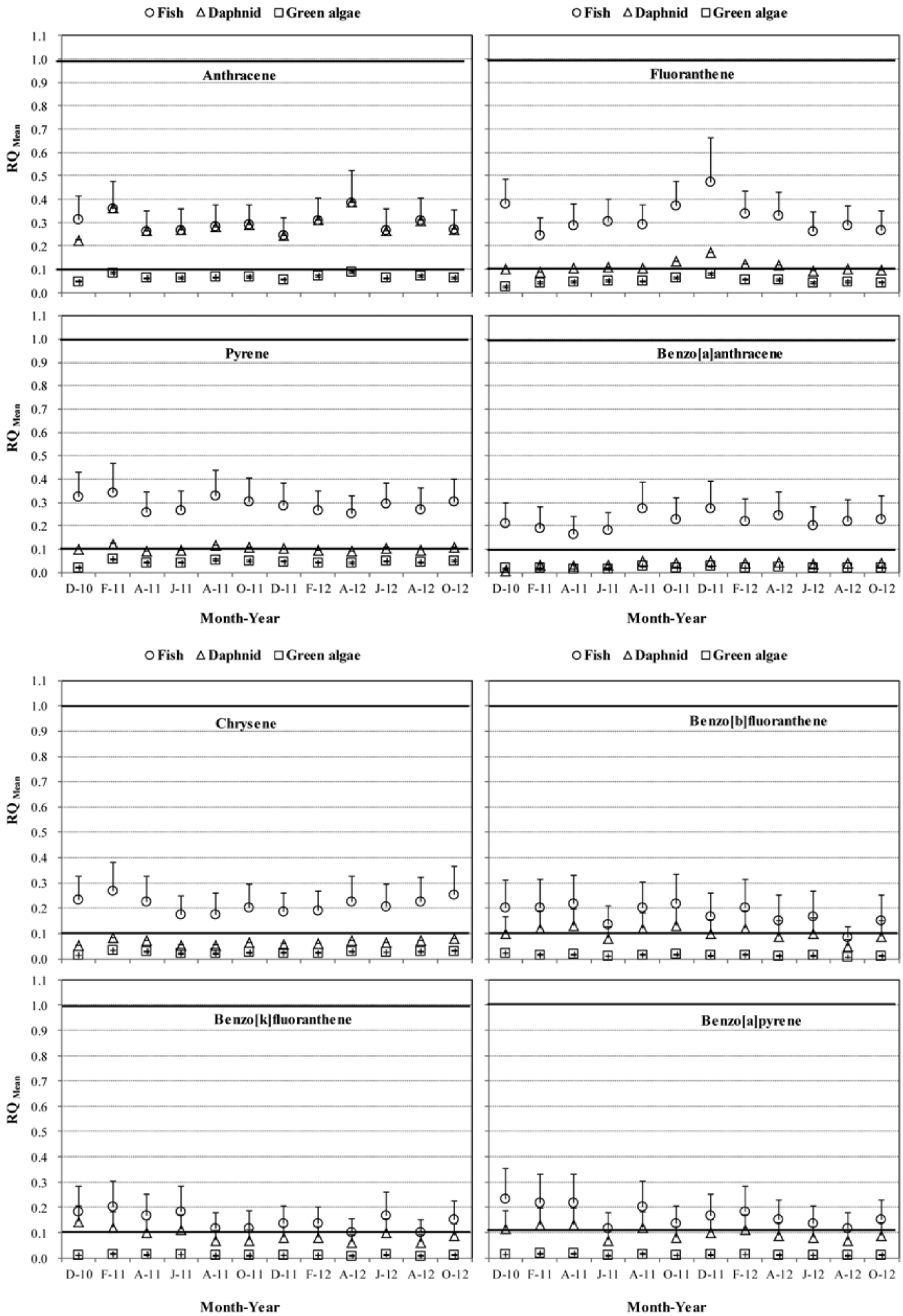

Fig. 4 Continued.

reference value, was proposed for the individual-value estimate (Solomon et al., 2000). In recent decades, some indicators and methods of different complexities have been proposed for the ERA of toxic chemicals in water (Qin et al., 2013). The potential for contamination to cause undesired environmental effects can be estimated from an index of ERA (Lee et al., 2008). Evaluation of green algae, daphnids, and fish has primarily been used in order to assess the potential aquatic ecotoxicological risks of the PAHs in the Dongjin River water system Korea. The individual RQ values of the investigated PAHs were calculated and are shown in Fig. 4. When a RQ value exceeded 1.0, it indicated that the pollutants in the area were of high risk, whereas RQ values of 0.01–0.1 and 0.1–1 equated to low and medium risks, respectively (Hernando et al., 2006). The RQ values for the 15 PAHs in the Dongjin River water system were below 0.01–0.1, indicating little risk to the relevant sensitive aquatic organisms, including green algae and daphnids, posed by the target compounds. In particular, the RQ values of most of the PAHs exceeded 0.1 for fish in all of the seasons at most of the sampling sites, which indicated that the fish were exposed to medium risk. Among the 15 PAHs, the RQ values of the low molecular weight 3-ring PAHs showed relatively higher RQ values than the medium/high molecular weight 4-, 5-, and 6-ring PAHs. Although such risk assessment is a useful exercise, the results should be treated with caution as the toxicity data were obtained from the ECOSAR model, which might be simplified. In addition, single-compound exposure scenarios are unrealistic in the real environment due to the mixture effect of multiple contaminants (Leung et al., 2012) which may cause considerable ecological concerns (Yan et al., 2013).

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