



Occurrence, Distribution and Risk Assessment of PAHs in Road Dust Sediment in Daejeon Metropolitan City, South Korea

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ABSTRACT

In this study, the occurrence and distribution of polycyclic aromatic hydrocarbons (PAHs) in road dust sediment (RDS) deposited from main traffic roads as well as residential and industrial areas were examined with a total of 144 samples in seasons. Based on the results, the concentrations of PAHs were found to be higher in winter and ranged between 0.15 and 1.94 mg/kg with an average of 0.65 mg/kg. The results indicate that pyrene, fluoranthene, phenanthrene, and benzo(ghi)perylene were commonly found in RDS, likely originated from vehicle exhaust emissions and fuel combustion by the diagnostic ratio study. Non-cancer risk from PAHs for humans exposed to RSD was found to be low (HI 1.2E-05). The estimated excess cancer risks by PAHs in RDS was also much less than the regulatory guideline for cancer risk (1×10^{-6}). Thus, current concentrations of PAHs in RDS are expected to be pose no threat to humans under the assumed exposure scenarios with applied exposure factors. However, more extensive and long-term sample analysis for gaseous and particulate PAHs is still warranted to determine potential risks to humans and the fate in the environment, since their concentration and distribution can be influenced by micro-metrological conditions, traffic volumes, or road clean-up conditions.

Keywords: distribution, risk assessment, road dust sediment, PAHs

1. Introduction

Potential health risks caused by fine dust in air has become a major issue of concern in Asian countries [1-4]. Road dust or street dust sediment from rapid urbanization is one of many sources of fine dust [5, 6]. Many countries make various efforts to reduce environmental pollution and its public health risks by monitoring road dust [7, 8], sweeping streets [9], applying sprinkling water on roads [10] and risk communication with public and industry. For example, in South Korea, when the fine dust concentration is higher than the regulatory limits (PM_{10} 100 $\mu\text{g}/\text{m}^3$ per day), the Korean Government takes a number of strict measures, including restricting the emissions from industrial facilities such as oil refin-

ery, coal-fired power plants and steel manufacturing facilities, enforcing the restricted driving with diesel vehicles in cities, and communicating risks with public to recommend limiting outdoor activities [11].

Pollutants emitted from various sources such as vehicles, industrial complexes, residential heating system, and coal-fired power plants can be accumulated directly on urban roads, mainly on roadsides, surrounding soils, and buildings by dry and wet-deposition [5, 12]. Sediment accumulated around roads is often called road dust sediment (RDS), or street dust residues. It may contain many organic pollutants (e.g., polycyclic aromatic hydrocarbons (PAHs), polybrominated diphenyl ethers, phthalates) [3, 13-16] and heavy metals [17, 18]. It can be introduced



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into wastewater treatment plants, rivers or other watersheds directly from the roads during rain, eventually impacting ecosystems in the environment [19].

As commonly detected pollutants in RDS, PAHs are often formed during the incomplete combustion of organic matters such as wood, coal, or oil (e.g., vehicle exhaust, electricity generating power plant, and manufacturing process) [13, 20, 21]. Emitted PAHs could be wet-precipitated into the soil by precipitation, adsorbed to atmospheric suspended dust, and introduced into the surface in a gaseous form [18, 21]. PAHs deposited on solid particles on the road surface can be re-scattered due to vehicle traffic by the wind and can cause potential hazards to humans through exposure routes such as inhalation, unintended ingestion, and dermal contact [22-25]. In particular, benzo(a)pyrene is known as “carcinogenic to humans” based on strong and consistent evidence in animals and humans [26]. The International Agency for Research on Cancer (IARC) also classifies benzo(a)pyrene into Group 1, a class of carcinogenic to humans [27]. In South Korea, due to potential toxicity of PAHs, they are designated as a group among toxic substances in air and the concentration levels of 16 PAH compounds are routinely monitored by air pollution monitoring network system [28].

According to previous studies, the concentration of PAHs is often high due to combustion of petrochemical fuels, diesel and gasoline-powered vehicles and highly affected by traffic [13, 16, 18, 20, 29]. By type of land use, it was common to show high concentrations in the industrial area and traffic roads rather than in rural area [15, 30, 31]. Previous studies in UK [32], Hong Kong [33], Brazil [34], Saudi Arabia [35], and China [36, 37] investigated potential carcinogenic hazards from PAHs in RDS. PAHs in RDS were commonly found to be higher concentrations in case of more road vehicles and higher carcinogenic risks in urban areas than in rural areas. Meanwhile, Hussain *et al.* [18] found that the concentration of PAHs shows different distribution depending on the season. Lim *et al.* [38] also revealed that the concentration distribution of particulate PAHs in the atmosphere displayed seasonal characteristics in South Korea. They reported that the risk of PAHs in the atmosphere tends to be determined by PAHs in particulate form rather than PAHs in gaseous type, and the concentration was higher in winter than in summer. While several studies on concentration levels and distribution characteristics of PAHs in RDS have been conducted [15, 39, 40], limited previous studies have quantitatively evaluated chemical characterization of PAHs in season and land use type and their potential risks to humans by PAHs in RDS.

In this study, 16 Priority PAHs were selected as target substances in RDS collected from Daejeon Metropolitan City (DMC) in South Korea. For monitoring the concentration levels of the PAHs and their distribution characteristics, 12 locations representing traffic roads, residential area roads, and industrial area roads were selected. The source of the chemicals was evaluated by using the diagnostic ratios of specific PAH species. In addition, seasonal variation (e.g., winter, spring, summer, autumn) for the PAHs concentration were examined. Based on the results, risk assessment according to potential exposure pathways was performed to determine excess cancer risk and non-cancer risk. The results of chemical characterization and risk assessment of PAHs can be used for providing the risk

communication with general public, policy makers and risk managers, and developing appropriate management options of RDS, if needed.

2. Materials and methods

2.1. Sampling Collection

In this study, a total of 12 sampling locations (144 samples) from three local districts (Daedeok-gu, Seo-gu, and Yuseong-gu) in DMC with approximately 1.5 million populations, one of the six major cities in South Korea, were selected to represent the roads from high traffic (T1~T4), residential area (R1~R6), and industrial complex (I1~I2), as shown in Fig. 1. Four main traffic roads were chosen, based on heavy traffic volume and speed from Traffic Data Warehouse System [41]. No traffic volumes for industrial complex are available in the system, however. The industrial areas were selected to identify the effect of PAHs emissions from industrial manufacturing processes and activities. The industrial complex currently occupies area of 2.3 million m² and consists of approximately 300 manufacturing facilities (e.g., machinery, electronics, chemicals, textiles, food, paper), recycling plants, and incineration facilities. Road dust sediment from the streets from residential areas with relatively high population density were examined. The residential areas were mostly located at least 2 km away from main traffic roads and industrial complex. The areas consist of many apartment complexes where people go by walks, rather than moving by heavy vehicle traffic. Table S1 presents the sampling locations and traffic volumes of the sites.

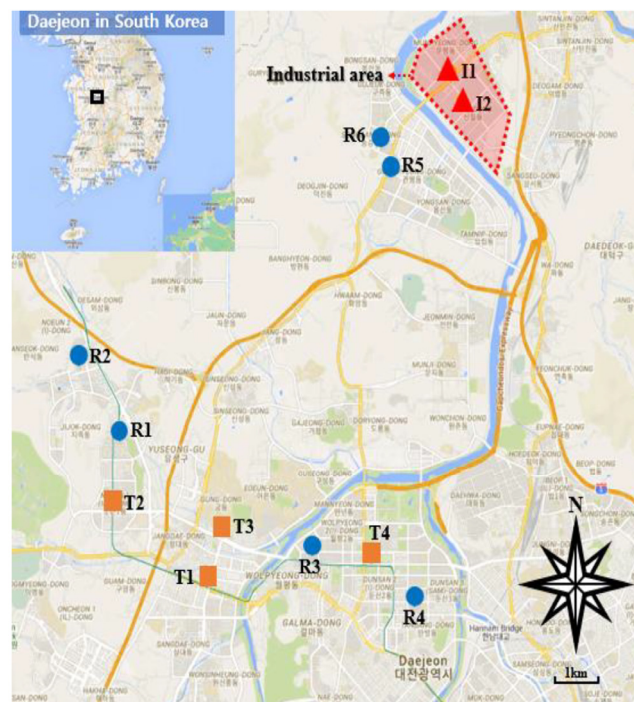


Fig. 1 Sampling sites of road dust sediment in DMC.

RDS samples were collected using a brush and a plastic dustpan at each site by visually observing the roads where the sediment was accumulated. Approximately 50 g of the collected samples in the dustpan was transferred into a 120 mL-glass bottle after removing large materials (e.g., twigs, branches, and gravels). Each sample was collected at 3 different locations within a radius of 50 m and blended in the bottle. The composite samples were immediately stored with an ice cooler and were then transported to the laboratory without being exposed to sunlight prior to analysis. Repeated sampling was conducted in each season (spring, summer, autumn and winter) at the same sites. The samples were screened and sieved into eight different fractions by particle size (<25 μm , 25-63 μm , 63-100 μm , 100-125 μm , 125-250 μm , 250-500 μm , 500-1,000 μm) in accordance with ASTM D6913. Previous studies reported that many pollutants in RDS can be found in the smaller size fraction (0~300 μm range) [42-44], since the particle size decreases, the specific surface area increases. In this study, the particle size fractions of RDS consisted of 0~250 μm (on average 70.9% for industrial area, 70.5% for high traffic roads, and 56.7% for residential areas).

2.2. Chemical Analysis for PAHs in RDS

A total 144 samples (consisted of 36 samples per each season) were analyzed for 16 PAHs targeted in this study, including Naphthalene (Nap), Acenaphthylene (AcPy), Acenaphthene (AcP), Fluorene (Flu), Phenanthrene (PhA), Anthracene (Ant), Fluoranthene (FluA), Pyrene (Pyr), Benzo(a)anthracene (BaA), Benzo(b)fluoranthene (BbF), Benzo(k)fluoranthene (BkF), Benzo(a)pyrene (BaP), Dibenz(a,h)anthracene (DbA), Chrysene (Chr), Benzo(ghi)perylene (Bghip), and Indeno(1,2,3-cd)pyrene (InP).

PAHs in RDS were analyzed by using GC/MS (GC 7890B and MS 5977A, Agilent Technologies) equipped with DB-5MS (Agilent Technologies, 60 m x 0.25 mm x 0.25 μm) after extraction and purification by referring to the benzo(a)pyrene analysis method [45]. After allowing the sample stored in the refrigerator to cool at room temperature, impurities such as large particles (>2 mm diameter) were removed, and approximately 2 g from the sample with less than 2 mm particle size was taken into a stoppered Erlenmeyer flask (250 mL). 80 μL of internal standard substances (Naphthalene-d8, phenanthrene-d10, pyrene-d10, benzo(a)pyrene-d12) (10 mg/L in methanol (MeOH)) was added to the Erlenmeyer flask. Then, about 5 g of anhydrous sodium sulfate was added into the Erlenmeyer flask with 10 mL of methylene chloride (MC):hexane (1:1 by volume). Extraction was carried out by sonication for 30 minutes using a sonicator, and then a supernatant was separated by a centrifuge (FLETA 5 PLUS, Hanil Scientific INC.) at 2,500 rpm for five minutes. The extracted solution was concentrated to 0.9 mL using a nitrogen concentrator (XcelVap, Horizon Technology). Then, hexane was added to make the final volume to 1 mL, being filtered using a 0.2 μm membrane filter. Purification process was carried out by conditioning with 10 mL of MeOH, 10 mL of MC, and 13.5 mL of hexane using a Florisil cartridge (WAT043390, WATERS), loading 1 mL of the extraction solution, and then eluting with 14 mL of MC:hexane (1:1 by volume). 200 μL of t-butanol was added to the eluted solution. The eluted volume was concentrated using the nitrogen concentrator until

it became 200 μL . To measure the concentrations of PAHs in the pretreated samples by GC/MS, high purity helium was used as the carrier gas at a constant flow of 0.6 mL/min and 1 μL volume of the sample was injected at a splitless mode (purge on time: 0.75 min). The GC temperature of injector was 300°C. The temperature program of the oven was started at 70°C (held 2 min), ramped to 240°C at 20°C/min (held 2 min), further ramped to 290°C at 5°C/min (held 2 min), then finally ramped to 310°C at 3°C/min and held for 15 min. Mass spectrometer conditions were as follows: ionization mode at electron impact energy at 70 eV was set, interface temperature was 300°C, ion source temperature was set at 320°C, and quadrupole temperature was 150°C. The GC/MS was operated in the selective ion monitoring mode for the scan (solvent delay: 6.8 min).

Linear curve of each PAH was obtained by identifying the peak areas of nine concentration levels of PAHs: 0.5 $\mu\text{g/L}$, 1 $\mu\text{g/L}$, 2.5 $\mu\text{g/L}$, 5 $\mu\text{g/L}$, 10 $\mu\text{g/L}$, 25 $\mu\text{g/L}$, 50 $\mu\text{g/L}$, 100 $\mu\text{g/L}$, and 250 $\mu\text{g/L}$. The linear curve used in this study was adjusted by a corrected recovery rate, and the recovery rate was found to be a range of 80~100%. The linear regression coefficients (R^2) of the 16 analyzed PAHs ranged from 0.9932 to 0.9999. Before analyzing PAHs by the GC/MS, a surrogate spike mix including naphthalene-d8, phenanthrene-d10, pyrene-d10, chrysene-d12, and benzo(a)pyrene-d12 was added to the RDS extracts. The surrogate compounds included naphthalene-d8 for Nap; phenanthrene-d10 for AcPy, AcP, Flu, PhA, AnT, and FluA, pyrene-d10 for Pyr, BaA, and Chr, benzo(a)pyrene-d12 for BbF, BkF, BaP, InP, DbA, and Bghip. *The recovery rates for four surrogates were ranged from 70.4~77.1%, as shown in Table S2).*

Laboratory blanks, duplicates, sample spikes, and calibration check samples were included during chemical analysis for quality assurance and quality control (QA/QC). All glassware was washed with 1-N HNO₃ and thoroughly rinsed with de-ionized water prior to use. All reagents used were analytical grade. Duplicate samples were collected and run every eight samples in each batch. PAHs were not detected above the detection limit (0.5 $\mu\text{g/kg}$) for any of the blanks. Procedures for the determination of method detection limits (MDLs) for PAHs in RDS in this study were described with the values in Supplementary Materials (Text S1 & Table S3).

2.3. Risk Assessment for PAHs in RDS

Receptors from potential exposure pathways by inhalation by air, unintended ingestion, and dermal contact were considered as general adult residents near the sampling locations and children (under 9 years old) because they were relatively vulnerable to potential health risks. Total human exposure to PAHs was calculated by summing the exposures calculated through all of the pathways from the sediment in this study. Exposure equations (Eq 1~3) for each pathway are presented as following, by examining the existing risk assessment guidelines and previous studies [46, 47].

Inhalation route

$$ADD = \frac{C_s \times IR_d \times PEF \times EF \times ET \times ED}{BW \times AT \times 365} \quad (1)$$

Ingestion route

$$ADD = \frac{C_s \times IR_s \times EF \times ET \times ED \times CF \times 10^{-6}}{BW \times AT \times 365} \quad (2)$$

Dermal contact

$$ADD = \frac{C_s \times SA \times AF \times ABS \times EF \times ET \times ED \times CF \times 10^{-6}}{BW \times AT \times 365} \quad (3)$$

Where, ADD: average daily dose (mg/kg/day), Cs: TEQ-tPAHs concentration in soil (mg/kg),

IRa : inhalation rate (m³/hr), IRs: ingestion rate (mg/day), AT: average time (year),

EF : exposure frequency (day/year), ABS: Soil to skin absorption factor (unitless),

ED : exposure duration (year), PEF: soil to air particle emission factor (m³/kg),

AF : Soil to skin adhesion factor (mg/cm²), SA: surface area (cm²/event),

BW : body weight (kg), CF: conversion factor (day/hr), ET: exposure time (hr/day)

Excess lifetime cancer risk (ECR) is obtained by multiplying the Lifetime Average Daily Dose (LADD) by the slope factor (SF) in Eq. (4). *The term of "excess lifetime cancer risk" indicates the possible occurrence of cancer in excess of the natural background risk, resulting from a lifetime exposure to carcinogens* [48].

$$ECR = LADD \times SF \quad (4)$$

Non-carcinogenic risk was also calculated by summing all the ADD values for each type of PAHs divided by the reference dose. The hazard quotients (HQ) of target PAHs for non-carcinogenic effect and hazard index (HI) were estimated by using Eq. (5) and Eq. (6), respectively.

$$HQ_i = ADD_i / RfD_i \quad (5)$$

$$HI = \sum HQ \quad (6)$$

2.3.1. Exposure factors

Exposure factors for this study include exposure time (ET), exposure frequency (EF), exposure duration (ED), body weight (BW), inhalation rate by air (IRa), ingestion rate (IRs), particle emission factor (PEF), adherence factor (AF), absorption factor (ABS), and surface area (SA). The values for the factors applied to the exposure equations were adopted from standard exposure handbooks and previous studies and literature [49-53] and other studies [33, 47]. Since the major exposure targets for the risk assessment in this study are Koreans, we used the exposure factors developed by previous studies for risk assessment [52, 53]. The detailed exposure factors applied in this study can be found in Table S4.

2.3.2. Toxicity values

2.3.2.1. Slope factors and reference doses

To determine the carcinogenic risk and non-carcinogenic risk using the human exposure of PAHs in RDS, the slope factor (SF) and reference dose (RfD) of PAHs through dose-response evaluation were obtained from previous studies. RfD values for oral exposure

have been developed by U.S. EPA IRIS (integrated risk information system) [54]: naphthalene (0.02 mg/kg/day), anthracene (0.3 mg/kg/day), acenaphthene (0.06 mg/kg/day), acenaphthylene (0.06 mg/kg/day) fluoranthene (0.04 mg/kg/day), fluorene (0.04 mg/kg/day), phenanthrene (0.0071 mg/kg/day), pyrene (0.03 mg/kg/day), benzo(a)pyrene (0.0003 mg/kg/day) and benzo(g,h,i)perylene (0.03 mg/kg/day). No other reference doses exist for the remaining six PAHs. Thus, in this study, non-carcinogenic risk was calculated for only 10 PAHs with the known RfD values among the 16 PAHs studied. Slope factors for ingestion, dermal-adjusted, inhalation are 7.3, 25, and 3.85 (mg/kg/day)⁻¹, respectively [21, 55, 56].

2.3.2.2. Toxic equivalency factor (TEF)

If the toxic mechanisms of individual compounds are the same, such as PAHs, their exposure assessment should be conducted using toxic equivalency factors (TEF) based on the toxicity of the substance that can represent the compound [57]. In this study, in order to determine ECR, the toxicity value of benzo(a)pyrene, the most toxic PAH, was set to be 1.0. The TEQ-tPAHs (mg/kg) value obtained by multiplying each TEF (1.0~0.001) of 16 PAHs by the arithmetic mean of each PAHs [58] was used to perform the risk assessment in this study.

3. Results and Discussion

3.1. Concentrations of PAHs in RDS

3.1.1. Concentration distribution of PAHs in RDS

Table 1 shows the average concentration of 16 PAHs from RDS in DMC for all seasons and the sum of 16 PAHs (\sum PAHs). The highest concentration was 1,939 μ g/kg at I1 and the lowest was 145.2 μ g/kg at R2. Compared with the results of studies conducted in other cities in South Korea [15, 29, 39, 40], the levels in this study were similar to those of Gwangju Metropolitan City [15]. In addition, compared with the results of other countries such as China [59-62], India [18], Pakistan [63], UK [32], and Germany [16], we found that the concentration levels of PAHs from RDS in DMC were relatively low, as shown in Table 2. As for the concentrations by land use type, as in other studies [29, 61], the highest concentration was observed in the industrial area, followed by main traffic roads and residential area. In the case of T1 and T2, the concentration levels were higher than those of other main traffic areas, partly because such sampling sites are located at near the highway exit with heavy traffic volumes.

The distribution by type of PAHs from RDS in DMC shows that the major PAH compound was Pyr (20% of 16 PAHs concentration), followed by FluA(17%), PhA(15%) and Bghip(10%). Such distributions in order were similar to those of Busan [40], Incheon [39], and Gwangju [15] metropolitan cities in South Korea. Lee and Dong [64] reported that PhA and FluA were found to be from diesel engine cars, Bghip from gasoline engine cars, while FluA, Flu, Pyr, BbF and InP were likely from oil combustion in industrial facilities, and AnT and BkF from wood combustion. PAHs distribution by each site are shown in Fig. 2. Distribution of PAHs from RDS in DMC showed the similar trends regardless of land use type, although the individual fractions of 16 PAHs were slightly different depending on the site.

Table 1. Average Concentration of 16 PAHs from RDS in DMC (unit: $\mu\text{g}/\text{kg}$)

PAHs	R1	R2	R3	R4	R5	R6	T1	T2	T3	T4	I1	I2
NaP	12.60	7.69	22.44	12.68	18.74	15.06	102.06	50.41	14.66	20.18	47.02	109.76
AcPy	2.84	1.42	4.27	1.92	3.02	3.52	31.93	2.93	3.00	3.77	12.87	13.23
AcP	3.16	2.74	1.66	0.38	25.80	5.31	17.37	49.73	3.46	3.83	48.83	40.43
Flu	3.70	2.19	5.67	4.31	7.65	6.53	34.73	26.03	3.36	5.84	41.98	37.03
PhA	25.88	17.44	45.46	33.81	56.72	50.34	166.80	185.07	36.51	54.07	275.33	210.15
AnT	4.50	1.67	5.23	3.84	8.15	9.28	35.08	26.23	3.72	8.59	71.80	29.59
FluA	30.63	24.41	53.11	35.44	56.52	63.32	178.06	146.83	50.36	67.81	339.74	266.45
Pyr	39.90	30.43	69.36	50.92	72.79	76.36	176.15	123.02	68.47	83.03	423.29	351.21
BaA	5.31	3.58	11.92	8.17	14.17	14.27	47.77	35.95	7.94	16.48	96.63	42.51
Chr	9.30	7.22	19.03	14.35	18.94	17.40	47.06	41.63	15.15	23.14	113.10	56.18
BbF	11.77	9.59	26.21	18.83	26.01	23.78	55.13	52.59	19.54	31.03	123.94	93.35
BkF	3.07	2.42	6.88	5.08	6.90	7.66	19.00	17.92	5.08	7.73	36.12	24.33
BaP	7.75	6.07	14.43	9.36	14.98	17.05	45.16	40.90	12.98	18.48	94.84	61.15
InP	6.80	5.21	12.40	8.48	9.25	11.67	21.42	13.30	6.65	9.18	46.42	35.81
DbA	2.03	1.76	2.71	1.75	2.05	2.19	5.70	3.77	1.85	2.45	10.95	6.73
Bghip	26.06	21.34	45.41	37.16	35.40	45.28	84.26	30.87	37.73	39.05	156.30	195.64
Σ PAHs	195.31	145.16	346.20	246.46	377.09	369.03	1067.66	847.19	290.46	394.67	1939.15	1573.55

Table 2. Comparisons of the Concentration of 16 PAHs in RDS with other Cities

Country	Location	Type of land use	Concentration (mg/kg)	Major compounds	Reference
Korea	Daejeon	Urban, Industrial	0.15~1.94	Pyr, FluA, PhA	This study
	Busan	Urban	0.40~4.24	FluA, Pyr, PhA	[40]
	Incheon	Urban	0.60~4.52	Pyr, FluA, PhA	[39]
	Gwangju	Urban	0.14~1.74	Pyr, FluA, PhA	[15]
	Ulsan	Urban, Industrial	11.84~245.12	BkF, Pyr, BghiP	[29]
China	Shanghai	Urban	1.02~138.99	FluA, BbF, Bghip	[59]
	Guangzhou	Urban	0.84~12.30	FluA, Chr, BkF	[60]
	Beijing	Urban	3700 (mean)	FluA, Chr, PhA	[62]
	Jilin	Industrial	5.93~25.89	FluA, PhA, Pyr	[61]
	Harbin	Industrial	1.33~11.55	PhA, FluA, Pyr	[61]
	Daqing	Industrial	0.58~4.66	PhA, FluA, NaP	[61]
India	Guwahati	Urban	1.40~43.79	Nap, BaA, Chr	[18]
Pakistan	Rawalpindi, Islamabad	Urban	0.04~0.24	BaP, BbF, BkF	[63]
UK	Newcastle	Urban	0.60~46.00	Chr, FluA, Pyr	[32]
Australia	Gold Coast	Urban	0.15~1.22	Pyr, PhA, BghiP	[75]
Germany	Dresden	Urban	0.95~27.83	FluA, Pyr, PhA	[16]

3.1.2. Source identifications of PAHs in RDS with diagnostic ratios

Fig. 3 shows the distribution of the average ratios of 16 PAHs at sampling sites with respect to the number of benzene rings. The PAHs were divided to 2-ring (NaP), 3-ring (AcPy, AcP, Flu, PhA, AnT), 4-ring (FluA, Pyr, Chr, BaA), 5-ring (BbF, BkF, BaP, DbA) and 6-ring (InP, Bghip) based on the number of benzene rings. Molecular distributions were drawn by mean ratios of different ring types of 16 PAHs in RDS from the residential areas (R1~R6), the main traffic roads (T1~T4), and the industrial areas (I1~I2). The PAHs with 4-ring showed higher average ratios than any other types of PAHs in all sampling sites. Other researchers also reported that the 4-ring PAHs (FluA, Pyr, Chr, and BaA)

have been found as the substances commonly emitted from residential, industrial, and traffic areas [15, 39, 40]. For 2-ring PAH, the average ratios were 5 % in R1~6, 6 % in T1~4, and 5 % in I1~2, respectively. NaP, selected as representative 2-ring PAH, has been known to have high volatility (0.085 mmHg at 25 °C) [65]. These lower ratios can be commonly found in the literature [18, 29]. It might be attributed to possible loss by its high volatility upon RDS sampling. On the other hand, relatively low proportions were shown in 5 and 6-ring PAHs. The PAHs can be also expressed as LMW(Low molecular weight; 2-ring PAHs), MMW(Middle molecular weight; 3 and 4-ring PAHs), and HMW(High molecular weight; 5 and 6-ring PAHs) by their molecular weight. It shows

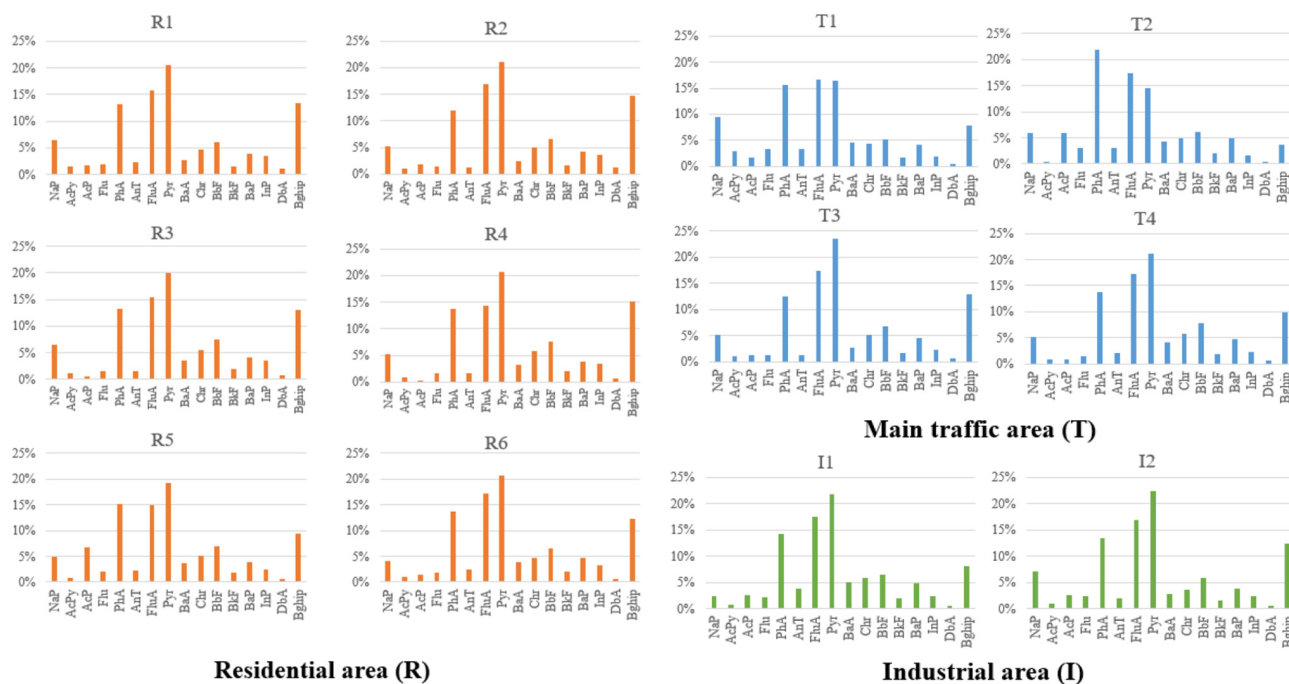


Fig. 2. 16 PAHs distribution from RDS in DMC by land-use type.

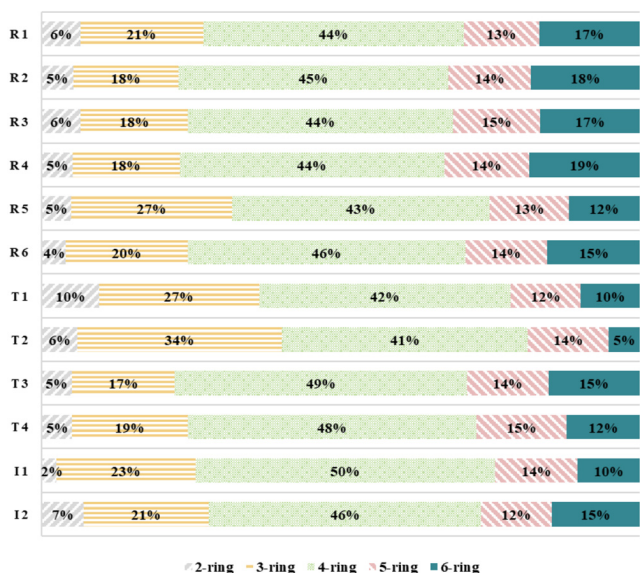


Fig. 3. Molecular distributions of 16 PAHs from RDS in DMC by sampling site.

the same distribution in order of MMW > HMW > LMW at all site. These findings suggest that the sources of PAHs are similar each other. HMW PAHs can be attributed to continuous emissions of vehicles moving and the accumulation of PAHs in limited dispersion of pollutants [64].

In order to identify potential emission sources of PAHs in RDS, six different diagnostic ratios (InP/(InP+BghiP), BaA/(BaA+Chr), $\sum 2\text{-}3\text{-ringPAHs}/\sum 4\text{-}6\text{-ringPAHs}$, AnT/(AnT+PhA), BaP/BghiP and FluA/(FluA+Pyr)) were calculated in this study (Fig. S1). The ratios

of InP/(InP+BghiP) with a value of 0.2 and BaA/(BaA+Chr) with greater than 0.2 indicate that there are also some possibilities of mixed sources such as wood or coal combustion and fossil fuel combustion. Such mixed sources of PAHs from fuel, biomass and coal combustion were probably due to the interaction between motorization, agricultural production (e.g., open burning of horticultural by-products) [64, 66, 67]. The ratios of $\sum 2\text{-}3\text{-ringPAHs}/\sum 4\text{-}6\text{-ringPAHs}$ with less than 1 and AnT/(AnT+PhA) with greater than 0.1 imply that the potential sources of PAHs in RDS samples of this study were caused by pyrogenic source rather than petrogenic source [68]. Yunker et al. [66] found that the ratio of FluA/(FluA+Pyr) is usually defined as a transition point of petrogenic to pyrogenic sources. The FluA/(FluA+Pyr) ratio with less than 0.5 indicates that the most sources were petrogenic, especially the range of 0.4-0.5 for fossil fuel combustion (vehicle emission). On the other hand, the ratio with greater than 0.5 implies that there were pyrogenic sources such as kerosene, grass, most coal and wood combustion [5]. Bjorseth [69] found that BghiP is mainly caused by traffic sources. The ratio of BaP/BghiP is known as an evidence that the major source of PAHs is a vehicle emission at value above 0.6 [70, 71].

As shown in Fig. 4 (a), both ratios at all sampling sites were spread over the region of the pyrogenic source. The main source of PAHs in the main traffic area, industrial area, and surrounding residential areas, when traffic is concentrated, can be affected by pyrogenic origin rather than petrogenic origin. The similar patterns can be observed in the FluA/(FluA+Pyr) ratio in Fig. 4(b). Among the pyrogenic area, it can be seen that a gathering appears at range of 0.4~0.5, which is close to the boundary of petrogenic value. Thus, fuel combustion such as vehicle exhaust is most likely a cause of PAHs. However, in the case of BaP/BghiP ratio, no similar

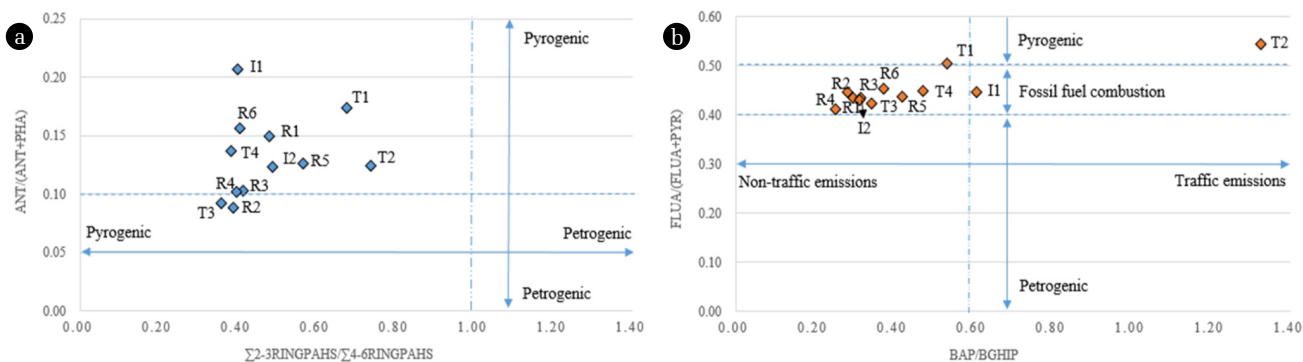


Fig. 4. Diagnostic ratios and potential sources of 16 PAHs from RDS in DMC.

pattern was observed. All data points except T2 are located in an area with less than 0.6, implying that here is non-traffic emission origin. T2 showed the highest diagnostic ratios of BaP/BghiP and FluA/(FluA+Pyr). The possible reason is related to a nearby highway exit and low vehicle speeds due to high traffic volumes. According to previous studies, Pandey et al. [72] found that the BaP/BghiP ratio in the area where known as clearly affected by traffic showed a value less than 0.6. Diagnostic ratios require caution when used to identify sources of PAHs from soil, sediment, and sludge rather than gaseous samples. When a clear and strong factor does not exist, a ratio different from the known value may appear [73]. Therefore, the BaP/BghiP ratio at points except T2 is expected to be a source of PAHs emission from other activities such as home heating or fuel combustion in industrial facilities in addition to traffic sources.

3.1.3. Seasonal variation of PAHs from RDS in DMC

Seasonal characteristics such as meteorological conditions can be a significant factor that may affect the distribution of PAHs in RDS. The atmospheric temperature, humidity and wind affected the pattern of conveyance of the PAHs [18, 72]. The concentration distributions of total PAHs (Σ PAHs) from the sampling sites by season are shown in [Fig. 5(a)]. The average concentrations of 16 PAHs by season were 0.61 mg/kg in autumn, 1.17 mg/kg in winter, 0.56 mg/kg in spring, and 0.26 mg/kg in summer. The average concentrations of 16 PAHs were highest in winter season about four times higher than in summer season, showing that strong sunlight and high temperature may promote the decomposition of PAHs. In the winter season, the temperature is relatively low and indoor and outdoor heating is actively supplied (Table S5). Therefore, the emissions through fuel combustion along with ambient temperature and limited dispersion conditions may have an effect on the PAHs concentrations in RDS. There is a high possibility that PAHs can be easily accumulated in RDS. The average concentration of PAHs in autumn was 1.41 mg/kg, 0.70 mg/kg and 0.28 mg/kg in industrial areas, main traffic roads and residential areas, respectively. The other seasons, PAHs concentrations in industrial areas and main traffic roads tend to be higher than in residential areas (Table S6). This means that, in the case of DMC, the target site of this study, PAHs generated from industrial activities and vehicle emissions are greater than the contribution caused by heating, or high-temperature cooking process of food in residential

areas. It is interestingly note that the average concentration of PAHs was high in R6 among residential areas in summer (0.57 mg/kg). This may be contributed to the geographical characteristics of the site close to the industrial complexes. For statistical analysis for seasonal variations, one-way analysis of variance (ANOVA) procedure for six sites (R3, R5, T1, T2, I1, and I2) was conducted to determine the differences of the concentrations. A seasonal difference of PAHs at 6 sites was statistically significant (p value < 0.05) at 95% confidence level with the highest concentration in winter (Table S7) This can be partly attributed to the influence of the meteorological conditions (e.g, low temperature, low solar radiation, and low photochemical degradation) and the effect of emissions by domestic heating in the cold season.

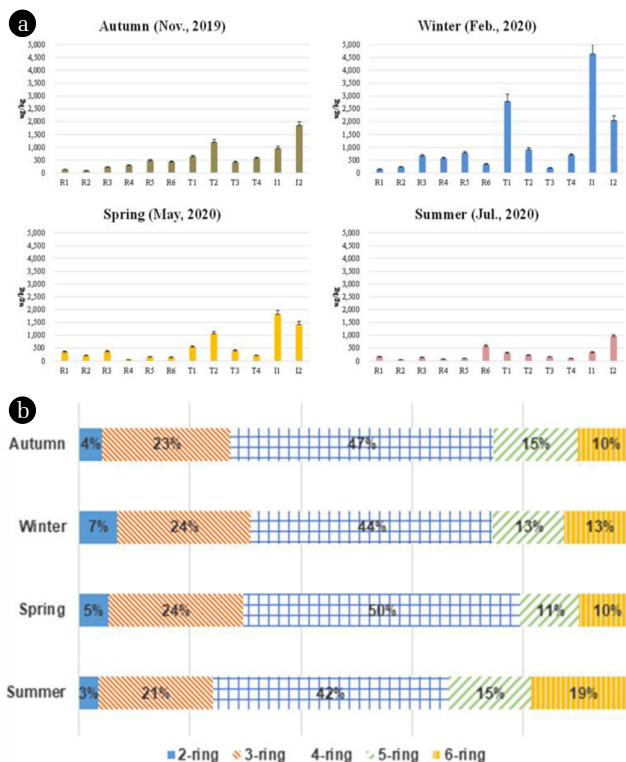


Fig. 5. Concentrations of 16 PAHs from RDS in DMC by season: by site (a) and by PAH ring (b).

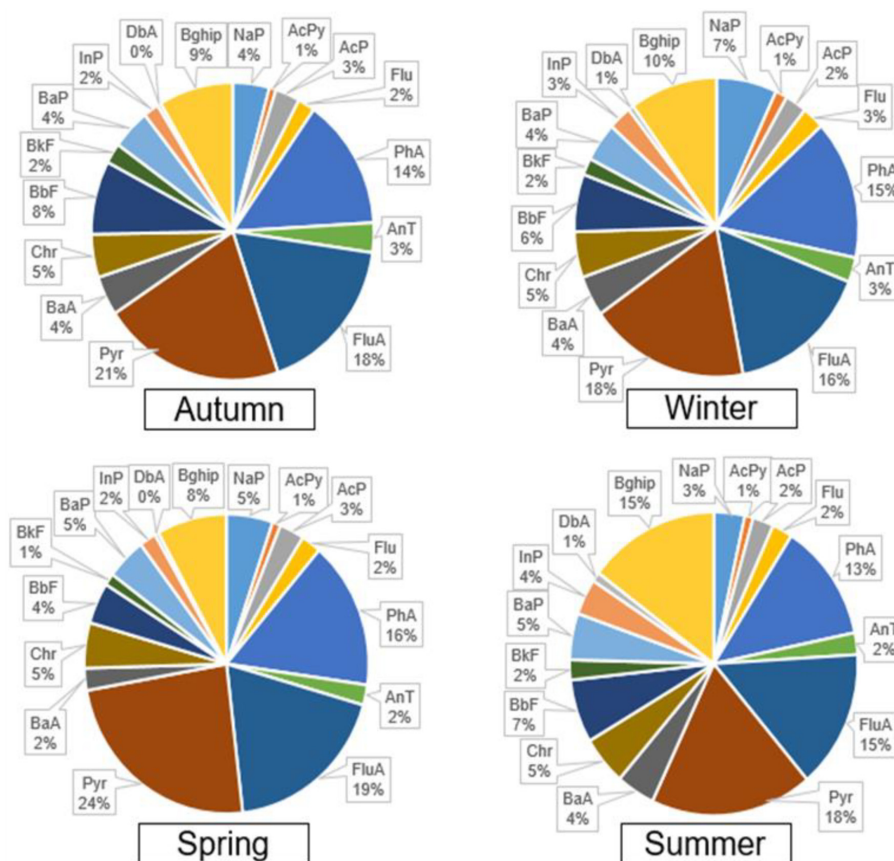


Fig. 6. Seasonal distribution of PAHs concentrations from RDS in DMC.

Fig. 5(b) displays the seasonal concentration distribution of PAHs by the number of benzene rings. In the summer when the lowest concentration level was found, the fraction of high boiling point compounds with 5-6 benzene rings was found to be relatively high. The summer in South Korea is relatively hot and humid between July and August with heavy rainfalls. Such climate conditions may be likely a cause of the fractional properties [18]. Especially, in the case of a 2-ring compound having a low molecular weight, it can be moved over a long distance and is susceptible to continuous deposition and volatilization [74]. However, as the compound with 4 benzene rings was most dominated than the others, there was no significant differences by seasons except for summer.

The fraction of individual PAHs by season is shown in Fig. 6. The major constituents commonly found in RDS include Pyr, FluA, PhA, and BghiP regardless of the season. Such compounds are characterized by having 4, 4, 3, and 6 benzene rings, respectively. When comparing compounds with the same number of benzene rings (e.g., Pyr and FluA), stability or individual physicochemical properties (e.g., boiling points of 404°C for Pyr and 448°C for Chr) are relatively similar. In the case of the compounds with three benzene rings and six benzene rings, there were no peculiarities by their structural and physicochemical properties. Therefore, it is considered that the results derived from the main constituent compounds by season in Fig. 6 are primarily due to the high concentrations of the four PAHs in the samples collected

from main traffic roads and roads in industrial area among the sampling sites (Table 1). Such compounds were also frequently found in street dusts from industrial areas and heavy traffic roads [16, 18, 20, 29, 75].

3.2. Results of quantitative risk assessment for PAHs in RDS

Hazard index (HI) values were calculated through deterministic risk assessment for the non-carcinogenic risk caused by PAHs from RDS in DMC (Table S8). Non-carcinogenic risk is evaluated as HI. If HI is greater than 1, it can be considered that potential non-carcinogenic risk may exist. According to the results of this study, all HI values for non-carcinogenic risk due to PAHs from RDS of DMC are much less than 1 for all sampling sites. Accordingly, it can be considered that there are no potential non-carcinogenic risks caused by PAHs in RDS.

The ECR of PAHs from RDS in DMC was calculated according to deterministic risk assessment (Table S9). Potential carcinogenic risks for both adults and children appeared to be very low with the exposure pathways in order of dermal contact, ingestion, and inhalation. Dermal contact is the exposure route that contributes the most among the risks. Among the sites, I1 site showed the highest cancer risks with the total ECR value of 6.32E-08 for adults and 6.67E-08 for children. Based the results of the cancer risk,

it was not likely to have potential cancer risks to humans because the calculated cancer risk values were much smaller than the regulatory limit of $1\text{E-}06$.

The level of ECR of PAHs from RDS in DMC was compared with the previous studies conducted in other cities [3, 75], although direct comparison of the risk results may be difficult due to different exposure assumptions and applied exposure factors. Gope et al. [3] presented the carcinogenic risks of PAHs from RDS in Durgapur in India and ranged from $2.4\text{E-}06$ to $3.8\text{E-}06$ for children and ranged from $2.1\text{E-}06$ to $3.4\text{E-}06$ for adults. Ma et al. [75] also reported the carcinogenic risks by ingestion, inhalation, or dermal-contact with urban road dust samples from 16 sites located in Australia were in the range of $6.25\text{E-}07$ to $4.41\text{E-}06$. However, the cancer risk levels estimated by this study were much lower than those of the previous results. The possible reasons could be attributed to the lower concentrations of PAHs found in RDS by this study as well as different exposure scenarios with the values of exposure factors applied. It can be concluded that both non-carcinogenic and carcinogenic risks to humans by potential exposures of PAHs in RDS in DMC may be expected to be significantly low under the assumed exposure scenarios with conservative and theoretical values of exposure factors.

4. Conclusions

In this study, the characteristics of the concentration and seasonal distribution of polycyclic aromatic hydrocarbons (PAHs) from road dust sediments (RDS) were examined, based on a total of 144 samples collected from 12 sites (traffic roads, residential and industrial areas) in Daejeon Metropolitan City (DMC) in South Korea. The concentrations of PAHs from RDS in DMC ranged from $145.2\ \mu\text{g}/\text{kg}$ to $1,939\ \mu\text{g}/\text{kg}$. In the case of the concentration of PAHs in RDS by land use type, industrial area was higher than main traffic roads and residential area. The PAHs species commonly found in the sediment include Pyr (20%), FluA (17%), Pha (15%), and BghiP (10%).

As a result of the emission source analysis, most of them are thought to be due to vehicle emissions. In terms of seasonal variation in PAHs concentrations in RDS, winter was the highest with an overall average of $1.17\ \text{mg}/\text{kg}$, while summer was the lowest with an overall average of $0.26\ \text{mg}/\text{kg}$, which is believed to be due to high temperature on the roads over summer likely promoted the decomposition of PAHs.

Based on the PAHs concentration data from RDS in DMC, the results of deterministic risk assessment (DRA) for adult and child showed that both non-carcinogenic and carcinogenic risks were significantly lower than the standard regulatory limits (Hazard Index=1.0 and cancer risk of 10^{-6}). Thus, potential human health risks caused by the PAHs in RDS by assumed exposure scenarios are expected to be relatively very low.

The merit of this study is that the characteristics and one-year seasonal distribution of PAHs compounds in RDS were examined with 144 collected samples from multiple sites. We were also able to identify potential sources of the chemicals in the road sediments with different land use types by using diagnostic ratios. The PAHs in the sediments would be influenced by a number of factors,

including source emission types, micro-meteorological conditions, road conditions with traffic volumes, and clean-up conditions by street sweepers. Thus, more extensive RDS sampling and analysis along with air monitoring data by different land-use types over a longer period of time would be beneficial to provide more clear and representative results by determining correlation and potential sources of PAHs between air and RDS. In addition, the risk assessment may need to be assessed in conjunction with the concentration of not only gaseous PAHs and but also particulate PAHs associated with PM_{2.5} in the atmosphere for potential risks of PAHs to human health.

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Conflict of interest statement

The authors declare that they have no conflict of interest.

Author Contributions

Conceptualization and study plan, Y.C.J. (Professor) and K.C. (Research Professor); methodology, G.L. (MEng.) and H.K. (MEng.); data organization and chemical analysis, H.S.S. (Professor), S.J. (Undergraduate student), B.K. (Master student), and Y.K. (Master student); writing review and editing, Y.C.J and K.C. All authors have read and agreed to the published version of the manuscript.

Appendix A. Supplementary Materials

The supplementary information provides summary of sampling locations, exposure factors, the climate characteristics of sampling sites in DMC, seasonal concentrations of PAHs by land use type, hazard index and excess cancer risks, and detailed diagnostic ratios of PAHs by sampling sites (Text S1, Table S1~S9, and Fig. S1).

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