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作成者:
メールアドレス:
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Some Results on Particulate Phase Polycyclic Aromatic Hydrocarbons (PAHs) in the Urban Atmosphere of Kumamoto

Hezhe Ji*, Daizhou Zhang, Ryoutai Shinohara, Wataru Fujiie, Minoru Koga, and Zongbo Shi

Faculty of Environmental and Symbiotic Sciences Prefectural University of Kumamoto, Kumamoto 862-8502, Japan,

*Email: g0575004@pu-kumamoto.ac.jp or voshi9192002@vahoo.co.jp

It has been well known that Polycyclic Aromatic Hydrocarbons (PAHs), which are mainly emitted from fossil fuel combustion and biomass burning, are significant species in polluted urban atmosphere because of their carcinogenic and mutagenic properties. A large number studies on PAHs have been carried out recently. However, the physical and chemical properties of PAHs in the atmosphere are yet well understood due to their complex variability in space and time.

In this study, suspended particulate matters were collected at two sites in Kumamoto, a city in southwestern Japan from December 2003 to May 2004. One site is in the central areas and is right besides a major road (hereafter as roadside site). The other site is on campus of the Prefectural University of Kumamoto (hereafter as residence site), which is located in a residential area with a distance of about 8 km from the center of the city.

Seven sets of total suspended particulates (TSP) samples were collected simultaneously at the two sites by using high volume air samplers. PG-60 complex membrane filters (Advantec Japan) were used to collect the samples. Sixteen PAHs (16 compounds specified on US EPA Method 610) in a mixture and surrogate, consisting of naphthalene- D_8 , phenanthrene- D_{10} , Pyrene- D_{10} , Chrysene- D_{12} , and Benzo(a)pyrene- D_{12} standards, were obtained from Ultra Scientific Inc (North Kingston, RI, USA). Surrogate PAHs were added to the samples prior to extraction. Every filter was cut into small pieces and the PAHs in the samples were extracted by a series of treatments for the preparation of their detection. The mass fractions of the PAHs were measured using a gas chromatography with mass selective detection.

A recovery test was performed on PAHs using the method described in Sienra et al. (2005). The average recoveries of 16 PAHs in four matrix spikes varied from 32% (naphthalene) to 106% (benzo[ghi]perylene). The average quantification limit was 0.002 ng m⁻³ and the detection limit was 0.00074 ng m⁻³. The PAHs concentrations were corrected for recovery efficiency during extraction and silica gel chromatography.

The concentration of the following compounds were determined in this study: Naphthalene (Nap), Acenaphthylene (Acy), Acenaphthene (Ace), Fluorene (Fl), Phenanthrene (Phe), Anthracene (Ant), Fluoranthene(Flu), Benzo(a)pyrene (BaP), Benzo(a)anthracene (BaA), Pyrene (Pyr), Benzo(b)fluoranthene (BbF), Benzo(k)fluoranthene (BkF), Chrysene (Chry), Indeno(1,2,3-cd)pyrene (IcdP), Dibenzo(a.h)anthracene (DahA), Benzo(g.h.i)perylene (BghiP).

The average mass concentrations of each PAH, measured and the average contribution of individual PAHs to total PAH masses at the two sites are listed in Table 1. The total concentration of the 16 PAHs at the roadside site was $10.08 \sim 99.12$ ng m⁻³ with its average about 46.60 ng m⁻³. At the residence site, the total concentration was $11.68 \sim 60.93$ ng m⁻³ with its average about 30.03 ng m⁻³. This is 54% higher than at the residential site. During the sampling period, the total concentration at the roadside site showed more extreme variations than that at the residence site, which can be expected and should be reasonable.

According to the average concentration of each PAH, Benzo(b)fluoranthene occupied the largest mass fraction in the samples at both sites (Table 1). Its concentration at the roadside site ranged from 3.5 to 36.84 ng m⁻³ with an average of 14.32 ng m⁻³, which accounted for 30.7% of the 16 PAHs. At the residence site, its concentration was in the ranged from 0.19 to 27.73 ng m⁻³ with an average of 9.05 ng m⁻³, It contributed 30.2% of the 16 PAHs. The second contributors in the detected PAHs at the two sites are different. At the roadside site, Indeno(1,2,3-cd)pyrene was in the second

place. Its concentration was $0.97 \sim 13.27$ ng m⁻³ with its average 5.31 ng m⁻³, which accounted for about 11.4% of the detected PAHs. At the residence site, Chrysene is in second place. Its concentration was $0.66 \sim 6.64$ ng m⁻³ with its average 3.20 ng m⁻³, which accounted for about 10.7% of the detected PAHs.

At the roadside site, PAHs with 2 and 3 rings (from Naphthalene to Anthracene in Table 1), which are more volatile than those with more rings, contributed $6.9 \sim 28.0\%$ of total detected PAHs in mass. PAHs with 4 rings (from Fluoranthene to Chrysene in Table 1) contributed $13.3 \sim 39.9\%$. PAHs with 5, 6, and 7 rings (from Benzo(b)fluoranthene to Indeno(1,2,3-cd)pyrene in Table 1) contributed $39.9 \sim 77.3\%$. In contract, at the residence site the relevant contributions are $4.1 \sim 21.6\%$, $23.1 \sim 55.1\%$, and $15.1 \sim 64.5\%$, respectively.

Benzo(a)pyrene-equivalent species, here including Benzo(a)anthracene, Benzo(b)fluoranthene, Benzo(k)fluoranthene, Benzo(g,h,i)perylene, Benzo(a)pyrene and Indeno[1,2,3-cd]pyrene, are chosen to show the carcinogenicity of the PAHs. At the roadside site, Benzo(a)pyrene-equivalent species contributed 70.4% of the detected PAHs and at the residence site 62.4%. However, the carcinogenicity index, calculated as in Yassaa et al. (2001), is from 0.88 to 8.59 ng m⁻³ at both sites with the average 2.87 at the roadside site and 2.95 at the residence site, suggesting the carcinogenicities at the two sites are not much different. Each Benzo(a)pyrene-equivalent species had a considerable contribution to the carcinogenicity at the roadside site while at the residence site Benzo(a)pyrene and Benzo(a)pyrene dominated the carcinogenicity.

Table 1
Mass concentrations of 16 individual PAHs and their contribution of total PAHs concentrations at the roadside and residential

PAHs	Residential Site		Roadside Site	
	Aver (range) (ng m ⁻³)	Contribution to total PAHs concentrations (%)	Aver (range) (ng m ⁻³)	Contribution to total PAHs concentrations (%)
Naphthalene	0.19 (0.02-0.49)	0.6	0.61 (0.06-1.53)	1.3
Acenaphthylene	0.06 (BDL-0.2)	0.2	0.31 (0.02-0.86)	0.7
Acenaphthene	1.61 (0.14-4.77)	5.4	3.2 (0.26-11.3)	6.9
Fluorene	0.11 (0.02-0.37)	0.4	0.18 (0.02-0.44)	0.4
Phenanthrene	0.07 (BDL-0.24)	0.2	0.06 (BDL-0.17)	0.1
Anthracene	1.28 (0.03-3.75)	4.2	2.04 (0.24-7.74)	4.4
Fluoranthene	2.58 (0.08-6.84)	8.6	2.1 (0.4 - 4.71)	4.5
Pyrene	2.2 (0.08-5.66)	7.3	2.14 (0.4 - 5.53)	4.6
Benzo(a)anthracene	1.92 (0.65-3.57)	6.4	3.9 (0.64 - 8.4)	8.4
Chrysene	3.2 (0.66-6.64)	10.7	3.16 (0.6 - 6.21)	6.8
Benzo(b)fluoranthene	9.05 (0.19-27.73)	30.2	14.32 (3.5 -36.84)	30.7
Benzo(k)fluoranthene	1.92 (0.05-5.49)	6.4	3.01 (0.73-7.76)	6.5
Benzo(a)pyrene	0.88 (0.02-3.88)	2.9	0.8 (0.18-1.79)	1.7
Benzo(g.h.i)perylene	1.4 (0.1-2.76)	4.7	4.53 (0.8 -12.6)	9.7
Dibenzo(a.h)anthracene	2.09 (0.29-6.14)	7	0.92 (0.18 -2.58)	2
Indeno(1,2,3-cd)pyrene	1.49 (0.13-2.87)	5	5.31 (0.97-13.27)	11.4
ΣΡΑΗ	30.03 (11.68-60.93)		46.6 (10.08-99.12)	

Note: 1. **\(\Sigma\)** PAH: total PAHs concentrations

2. BDL: below detetion limit.

Reference

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