PAHs in the aerosol of Higashi-Hiroshima, Japan

Polycyclic aromatic hydrocarbons (PAHs) in the aerosol of Higashi Hiroshima, Japan: pollution scenario and source identification

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Abstract. Polycyclic aromatic hydrocarbons (PAHs) are considered to be atmospheric pollutants and well-known human carcinogens. This study analyzes the pollution scenarios of PAHs in the aerosol of Higashi Hiroshima, Japan. We examined the characteristics, the influences of meteorological conditions, the distribution and seasonal variation of PAHs in aerosol samples collected from June 2000 to May 2001. The concentration of PAHs in the aerosol of Higashi Hiroshima was ranging from 11.8 μg/g to 157.5 μg/g with a mean concentration of 63.4 μg/g. Results showed that there is positive correlation between aerosol PAH concentrations and ambient temperature but a relatively little correlation with solar radiation. Seasonal variation was observed with a higher concentration during winter and lower concentration during summer. Apart from that, principal component analysis and molecular diagnostic ratios were also used to characterize and identify possible PAHs emission sources. Results obtained strongly suggested that vehicular emissions appeared to be the major source of aerosol PAHs in this study although other sources do have some degree of influence.

Keywords: Polycyclic aromatic hydrocarbons, seasonal variation, emission sources, factor analysis, vehicular emissions

1. Introduction

Polycyclic aromatic hydrocarbons (PAHs) are ubiquitous pollutants in the environment and are of major concern due to their well-known carcinogenic and mutagenic properties (Kameda et al., 2005). PAHs in the atmosphere mainly originate from incomplete combustion of fossil fuels such as petroleum, coal, wood and oil (Chang et al., 2006). PAHs were also detected in smoke from industry such as steel and iron factories (Tang et al., 2005).

PAHs can be present in the atmosphere as particulates or gasses. Gaseous PAHs usually contain more fractions of lower molecular weight PAHs while particulate PAHs contain more fractions of higher molecular weight and higher carcinogenicity., aerosols with a diameter of 10 μm or less are of major concern due to the fact that they can be inhaled into the deeper respiratory tract regions easily (Kameda et al., 2005). Thus, the particle-bound PAHs are considered to be the most hazardous substances to human health. From the view of health concern, studying the existence and behavior of particle-bound PAHs is extremely important. Therefore, this group of pollutants has received much attention in air pollution studies worldwide for decades.

The recently enacted or proposed regulation of PAHs in Europe and by the WHO (Air qualities guidelines for Europe, 2000; Bostrom et al., 2002) clearly reflects their potential health risk. In spite of this, PAHs have received relatively less attention in Japan compared with other developed nations. In Japan, no environmental standard for atmospheric PAHs has been established except for benzo(a)pyrene, which is self-controlled in each factory (Kameda et al., 2005). Basically, only a few studies of atmospheric PAHs had been carried out in Japan. Most of them focus on the metropolitan city of Tokyo or industrial areas, such as Fuji, as their sampling sites.

Ohura et al. (2004) studied the PAH concentration of two industrial areas in Japan; namely Fuji and Shimizu. They concluded that the origins of atmospheric PAHs in these sites were dominated not only by traffic but also by the stationary industrial sources such as power plants and paper-making plants. Kakimoto et al. (2002) had the similar conclusion on the atmospheric PAHs of Kitakyushu, another industrial area located at the south of Japan.

Kamimoto et al. (2001) studied the relationship between PAH concentration and several metal and inorganics ions in atmospheric samples collected from Kanazawa, Sapporo and Tokyo. They found that a strong correlation only occurred in the winter samples taken from Sapporo but not in the samples taken from Tokyo and Kanazawa.

Tang et al. (2005) studied particle PAHs in the Pan-Japan Sea countries. Seven cities in Japan, China, South Korea and Russia were selected as sampling sites. They found that samples from Kitakyushu seemed to be affected by coal stove emissions and samples from Tokyo and Sapporo were affected mostly by diesel vehicle emissions. Hayakawa et al. (2002) compared the PAHs and nitro-PAHs in the airborne particles of Kanazawa. The relative composition of PAHs and nitro-PAHs indicated that the samples were affected strongly by diesel exhaust particles.

In conclusion, the studies of particulate PAHs in Japan showed that traffic emissions were the major source of PAHs in urban areas and stationary sources made significant contributions to the PAHs in some industrial areas. However, these studies were limited to only a few cities and data collected still cannot be used to build a complete database to evaluate the pollution scenario of PAHs in the country. Undoubtedly, further studies covering other sampling areas and a wider study scope is essential for a better understanding of PAH pollution in the atmosphere of Japan.

The study of atmospheric PAHs in Hiroshima is relatively scarce. To our best understanding, to date only Ozaki et al. (2005) have published some preliminary data on the particulate PAH concentrations in Higashi Hiroshima, a suburban area of Hiroshima. However, further study on the samples with respect to PAH distribution, source or relationship to the meteorological conditions was not attempted. Thus, this study attempts to analyze the preliminary data of Ozaki et al. (2005) in order to have an overview of the pollution scenarios, distributions and possible sources of the particulate PAHs in Higashi Hiroshima.

2. Experimental

The detail of sampling and analytical methodology was described in Ozaki et al. (2005). In brief, the sampling of aerosol was carried out on the rooftop location of the Engineering Faculty of Hiroshima University during 6th June 2000 to 28th May 2001. The 8-storey building was located about 2.5 km from the town center with the nearest main road approximately 200 km away. Aerosol particles (with particle size less than 7 μm) were collected on 0.6 μm glass fiber filters (GB-100R, Advantec) by a high volume air sampler (HVS-500-5, Shibata) operated at a constant flow rate of ~0.5 m^3/min. Each sample was collected over a period of 24 hours. Samples collected were dried in a desiccator for 2 days. Our study focused on the aerosol particles with particle size less than 7 μm due to the fact that this portion of atmospheric particles includes fine and ultrafine particles which could be inhaled and deposited deep in the human respiratory system (Kameda et al., 2005). Altogether 19 samples had been collected throughout the sampling period as mentioned above.

Samples were then extracted ultrasonically with 10.0 ml of dichloromethane for 60 minutes. The recovery of the extraction was in the range of 50 to 80%. The extracts were then concentrated under a gentle nitrogen flow to 2 ml and subjected to GC-MS analysis to quantify 13 PAHs: acenaphthylene(Ace), acenaphthene (Act), fluorene (Flu), phenanthrene (Phe), anthracene (Ant), fluoranthene (Flt), pyrene (Pyr), benzo(a)anthracene (BaA), chrysene (Chr), benzo(b)fluoranthene (BbF), benzo(k)fluoranthene (BkF), benzo(e)pyrene (BeP) and benzo(a)pyrene (BaP).

Meteorological data were obtained from the website of the meteorological monitoring station in the botanical garden of Hiroshima University. (http://home.hiroshima-u.ac.jp/hirodas/). The sampling site of this study is about 380 m away from the meteorological observation station.

Factor analysis, namely principal component analysis (PCA) had been carried out in this study. It was performed by SPSS 11.5 software package. Source groupings were determined using PCA with varimax rotation and retention of principle components having eigenvalues greater than 1 of the complete data set of PAH concentrations.

3. Results and discussion

3.1 POLLUTION SCENARIO

Figure 1 illustrates the PAH concentration in the aerosol of Higashi Hiroshima from June 2000 to May 2001. The concentration of PAHs in the samples analyzed ranged from 11.8 to 157.5 μg/g aerosol with a mean concentration of 63.4 μg/g. As shown by figure 1, PAHs with 4 rings and 5 rings are predominant in the aerosol samples. This is in good agreement with the theoretical fact that the higher molecular weight PAHs are generally less volatile and attached to the particle phase while the reverse is true for the lower molecular weight PAHs (Fang et al., 2004).

Figure 1 indicates a clear seasonal variation whereby the concentration of PAHs was higher in the winter (December – February) and lower in the summer (June – August). This result is consistent with the findings of many other researchers worldwide (Tang et al., 2005; Ohura et al., 2004). In general, it is known that atmospheric PAH concentrations are elevated in winter as compared those in summer. It is believed that the concentration is dependent on several seasonal factors and atmospheric loss mechanisms such as photolysis, radical reactions and so on (Ohura et al., 2004). Therefore, the lower concentration in summer could be accounted for by the established theory.

As shown in Table I, traffic emissions appeared to be the main source of particulate PAHs in Japanese cities. For industrialized cities such as Fuji, the particulate PAH concentrations are much higher than in mega-cities like Tokyo and Sapporo due to the contribution from the industrial emissions. The average concentration of PAHs in the aerosol of Higashi Hiroshima is 2.39 ng/m^3. This value is lower than those measured in all Japanese cities listed in table 1, indicating that the atmosphere in Higashi Hiroshima is relatively less polluted when compared to other Japanese cities. Generally, in Japan, PAHs mainly come from anthropogenic sources such as fossil fuel combustion and industry emission (Tang et al., 2005). As a suburban area, Higashi Hiroshima has relatively low population and less anthropogenic activities compared to other sampling cites listed above, resulting in a cleaner atmospheric environment in terms of PAH pollution.

However, comparing our study with some other studies worldwide, the concentration of PAHs in the aerosol of Higashi Hiroshima is higher than those measured in the urban areas in the USA and Brazil (Table I). In other words, the atmosphere of Higashi Hiroshima, a suburban area in Japan, is more polluted with

PAHs than some urban areas in other countries. This might indicate that the overall PAH pollution in the atmosphere of Japan is relatively higher compared to some other countries worldwide.

Among all PAHs, BaP is the most investigated PAHs and often used as an indicator of the total PAHs. Figure 2 illustrates the BaP distribution in the aerosol samples. More than 50% of samples had a BaP concentration ranging from 0.1 to 0.2 ng/m^3. However, about 5% of samples, those taken in the winter, had relatively high concentrations of BaP ranging from 0.9 to 1 ng/m^3.

Table II shows the concentration of BaP in the aerosol of Japanese cities and other countries. The average BaP concentration in this study is 0.3 ng/m^3. This value is much lower than the annual average BaP concentration in particulate samples taken from a busy traffic roadside in Tokyo (0.63 ng/m^3), Sapporo (0.52 ng/m^3) and Kitakyushu (0.61 ng/m^3) (Kamimoto et al., 2001). When compared with the average concentration of BaP in other studies, the BaP concentration of this study is lower than those measured in Seoul, Korea but higher than those measured in Baltimore, USA and Sao Paulo, Brazil. Higashi Hiroshima also has a much higher aerosol bounded BaP concentration as compared to New Brunswick, a suburban area in USA.

Based on the total PAH concentration (Table I), the atmosphere in Higashi

Hiroshima seems to be relatively less polluted compared with other Japanese cities. However, when compared with other studies worldwide, it seems that the atmospheric pollution of PAHs in Higashi Hiroshima is relatively more severe than some urban areas of other countries. Therefore, setting up a systematic environmental standard for PAHs is crucial in order to improve the air quality. Since the carcinogenicity of PAHs varies from one species to another, the actual health impact of the polluted atmosphere is determined by the composition of the bound PAHs as well as by the total PAH concentration. As a documented carcinogenic PAH, the relatively high loading of BaP in winter might indicate the necessity to further investigate the total toxic effect of the aerosol in Higashi Hiroshima.

3.2 THE ROLE OF METEOROLOGICAL CONDITIONS

The concentration of PAHs in the atmosphere depends on the source/sink mechanism in the atmosphere which is greatly influenced by the meteorological conditions under which the emissions were released. The ambient temperature affects the gas/particle partitioning of PAHs while the solar intensity influences photodegradation. Apart from that, wind speed and wind direction are the key factors in determining the concentration of pollutants due to long term transport. Many researchers found a good correlation between PAH concentration and meteorological parameters (Panther et al., 1999; Fang et al., 2004).

Figure 3(a&b) shows the correlation between PAHs and ambient temperature as well as solar intensity in this study. PAH concentration is observed to have a good correlation with ambient temperature (R^2=0.72) but relatively a poor correlation with solar intensity (R^2=0.58). This result is consistent with the findings of Fang et al., 2004, suggesting that photodegradation might be relatively less significant compared with gas-particle partitioning in governing the particulate PAH concentration in these sampling sites.

Generally, photochemical transformation has been considered to be the most important mode of atmospheric decomposition of both particulate and gas phase PAHs (Chetwittayachan et al., 2002). The impact of sunlight on particulate PAHs such as Flt, Pyr and BaA has been investigated and found that they rapidly decayed at the beginning of the experiment (1 hour to several hours) and then appeared to be constant (Kamens et al., 1988; Fan et al., 1995). This might partly explain the poor correlation observed between the aerosols PAH concentrations and solar intensity. Apart from that, it was found that relative humidity, wind speed and wind direction had no significant impact on PAH concentrations in the aerosols.

3.3 INDIVIDUAL PAH DISTRIBUTION AND POSSIBLE SOURCES

Figure 4 shows the mean distribution percentage of each PAHs in the aerosol samples. Among all PAHs species, BbF, BaP, Chr and Flt appeared to be the most abundant PAH species in the samples while the contribution of most of the 3 ring PAHs was minimal except for Phe. These highly abundant PAH species strongly suggested that vehicular emissions are the possible source due to the fact that these species had been well-documented to be found in vehicular emissions including gasoline and diesel exhaust. (Panther et al., 1999; Fang et al., 2004)

Molecular diagnosis ratios have been the most frequently used tools for the source identification of atmospheric PAHs although its reliability is still widely debated. Table III presents the molecular diagnosis ratios in this study compared with the values of other researchers.

The BaA/Chr ratio in this study is 0.51. This value falls within the range of those from vehicular emissions particularly gasoline emissions. It is also similar to those measured from used motor oil emissions. In addition, the BaA/Chr ratio is used as an indicator of the aging of atmospheric aerosol with higher values indicating freshly emitted air masses (Kakimato el al., 2002). The BaA/Chr ratio in this study (0.51) is lower than those obtained from Tokyo, Sapporo and Kitakyushu which ranged from 0.69 to 0.77 (Kakimato el al., 2002), suggesting that the aerosol samples in Higashi Hiroshima are aged compared to these cities.

The mean BeP/BaP ratio is 0.80 which is obviously not within the range of all documented sources but closest to those of gasoline emissions. It has a higher emission of BaP than gasoline emissions. Many studies indicated that biomass burning and some industry stationary sources such as steel industry have a relatively higher emission factor for BaP compared with BeP (Jenkin et al., 1996; Yang et al., 1998). This implies that the aerosol samples probably were affected by some other sources that had higher emissions of BaP than BeP.

The ratio of Pyr/Bap in this study is 1.00 that is fairly close to those from gasoline and diesel emissions. On the other hand, the Chr/Bep ratio of this study is 1.95 which falls in between the value of gasoline emissions and diesel emissions, suggesting that vehicular emissions (a mixture of gasoline and diesel emissions) appeared to be the major source.

The molecular diagnosis ratio strongly suggested that vehicular emissions (gasoline and diesel) are the major source of the aerosol PAHs with other sources possibly giving some degree of contribution. In order to further analyze the possible sources of the aerosol PAHs, Principal Component Analysis (PCA) was used. PCA is the oldest and most widely used multivariate statistical technique in atmospheric sciences.

Table IV provides the results of PCA in this study. The results showed that three factors explain the main part of the data variance; therefore, one element for each factor can be chosen as a tracer. Factor I explained 69.3% of the total variance with high loading for almost all PAHs determined, particularly Ace (0.914), Ant (0.973), Flt (0.920), Pyr (0.948), Chr (0.955) BkF (0.866), BaP (0.840) and BbF (0.920). In this study, Ace was chosen as the tracer of vehicular emissions as suggested by some other researchers (Simcik et al., 1999). There have been numerous studies focused on the predominant PAHs species emitted by various sources including vehicles and industry. Chang et al. (2006) and Yang et al. (1998) concluded that Ace, Flt, Pyr and Chr are the predominant PAH species in diesel vehicle emissions while Izumikawa et al. (1989) found that BkF, BbF, Pyr and BaP are the major PAH species in gasoline emissions. Venkataraman (2000) also found Chr, benzo(ghi)perylene, coronene and Ant in gasoline emissions. Factor 1 seemed to fit the criteria of vehicular emissions very well. Therefore, it was selected to represent vehicular emissions (a mixture of gasoline and diesel emissions).

Factor 2 accounts for 13.5% of the total variance with moderate loading on Act

(0.655), which was found predominantly from stationary industrial sources and coke ovens (Yang et al., 1998; Simcik et al., 1999). Therefore, Factor 2 probably represents stationary industrial emissions nearby the sampling sites. Factor 3 has a moderate loading of BaA and Act accounting for 8.3% of the total variance. BaA was found to be a predominant species and tracer of natural gas combustion (Simcik et al., 1999; Bourotte et al, In press; Lee et al, 2006). It is believed that Factor 3 represents natural gas combustion emissions.

Based on PCA results, it can be concluded that the aerosol PAHs in Higashi Hiroshima are predominantly emitted by vehicular sources although other sources such as natural gas combustion do have some degree of influence.

4. Conclusion

The concentration of PAHs in the aerosols of Higashi Hiroshima ranged from 11.8 μg/g to 157.5 μg/g with a mean concentration of 63.4 μg/g. A clear seasonal variation was observed with a higher concentration in winter compared with those in summer. BbF was the most abundant PAH in the aerosol samples collected in Higashi Hiroshima. It was found that the total PAH concentration had a good correlation with ambient temperature, suggesting that gas-particle partitioning may be an important factor governing the aerosol PAH concentration. Molecular diagnosis ratios imply that vehicular emissions were the possible major pollutant sources in Higashi Hiroshima. Statistical analysis of the data further revealed that vehicular emissions are an important determinant of PAH concentration in Higashi Hiroshima.

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Figure Captions

Figure 1:

PAHs concentration in the aerosol of Higashi Hiroshima (Jun 2000 – May 2001)

Figure 2:

Benzo(a)pyrene distribution of the aerosol of Higashi Hiroshima (Jun 2000 – May 2001)

Figure 3:

Correlation of PAHs concentration with (a) Ambient temperature (b) Solar intensity

Figure 4:

Mean distribution of individual PAHs in the aerosol in Higashi Hiroshima (Jun 2000 – May 2001)

Table I

Comparison of mean concentration of particle PAHs with other studies in Japan and worldwide

Location	**Area**	**PAHs (ng/m^3)**	**Possible sources**	**Reference**	**No of PAHs determined**
Hiroshima	*suburban*	*2.39*	*traffic*	*This study*	13
Tokyo	metropolitan city	5.96	traffic	Kakimoto et al., 2002	10
Sapporo	urban	6.81	traffic	Kakimoto et al., 2002	10
Kitakyushu	Industrial	6.86	traffic, industry	Kakimoto et al., 2002	10
Fuji*	Industrial	365 (summer)	traffic, industry	Ohura et al., 2004	39
		468 (winter)	traffic, industry	Ohura et al., 2004	39
Shimizu*	Industrial	270 (summer)	traffic, industry	Ohura et al., 2004	39
		309 (winter)	traffic, industry	Ohura et al., 2004	39
Texas, USA	urban	1.89	-	Park et al., 2002	13
Sao Paulo, Brazil	urban	0.23	-	Dallarosa et al., 2005	12
Guangzhou, China	urban	39.4	-	Tan et al., 2006	11
SIRIM, Malaysia	rural	0.30	-	Omar et al., 2002	17
Finocalia, Greece	rural	0.2-2.0		Omar et al., 2002	-

*values include vapor and particulate phases

Table II

Comparison of mean concentration of particle BaP with other studies in Japan and worldwide

Location	Area	BaP (ng/m^3)	Reference
Hiroshima	*suburban*	*0.30*	*This study*
Tokyo	metropolitan city	0.63	Kakimoto et al., 2002
Sapporo	urban	0.52	Kakimoto et al., 2002
Kitakyushu	Industrial	0.61	Kakimoto et al., 2002
Seoul, Korea	urban	2.55	Lee et al, 2006
Baltimore, USA	urban	0.124	Lee et al, 2006
Sao Paulo, Brazil	urban	0.28	Lee et al, 2006
New Brunswick, USA	suburban	0.088	Lee et al, 2006

Table III

Molecular diagnosis ratios of aerosol PAHs

	This study	**Gasoline**	**Vehicular emission**	**Diesel**	**wood combustion**	**used motor oil**
BaA/Chr[a,b]	0.51	0.28-1.20	0.40-0.58	0.17-0.36	0.93	0.5
BeP/BaP[b]	0.80	1.1-1.3	-	2.0-2.5	0.44	-
Pyr/BaP[c]	1.00	0.85		0.81	0.71	-
Chr/Bep[d]	1.95	2.50		1.60	2.40	-

[a] Fang et al (2004)

[b] Bourotte et al (In press)

[c] Khalli et al (1995)

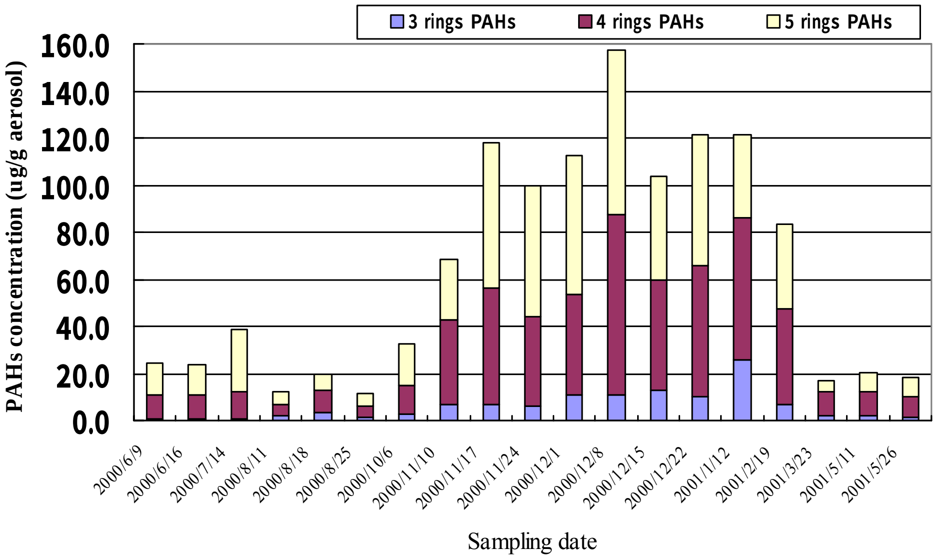
[d] Li and Kamens (1993)

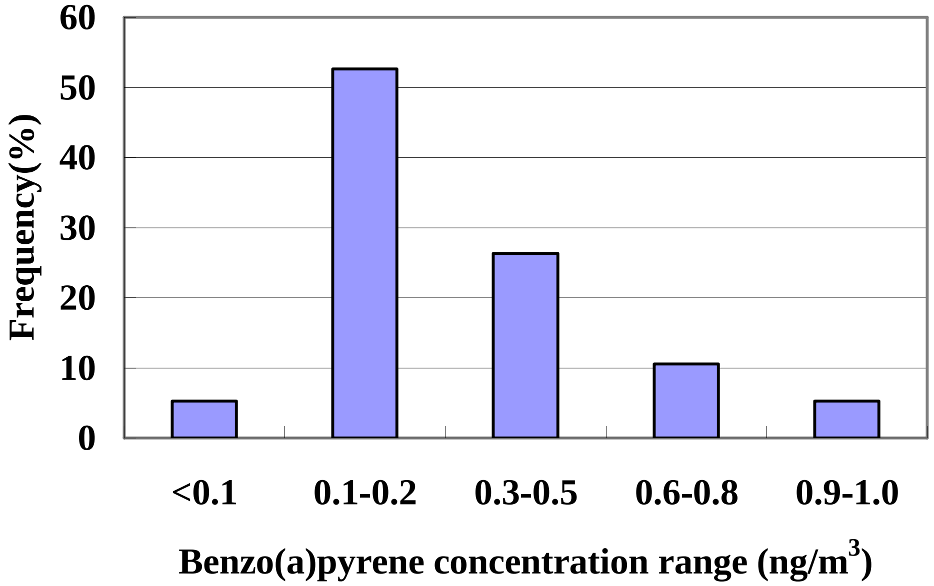
Table IV

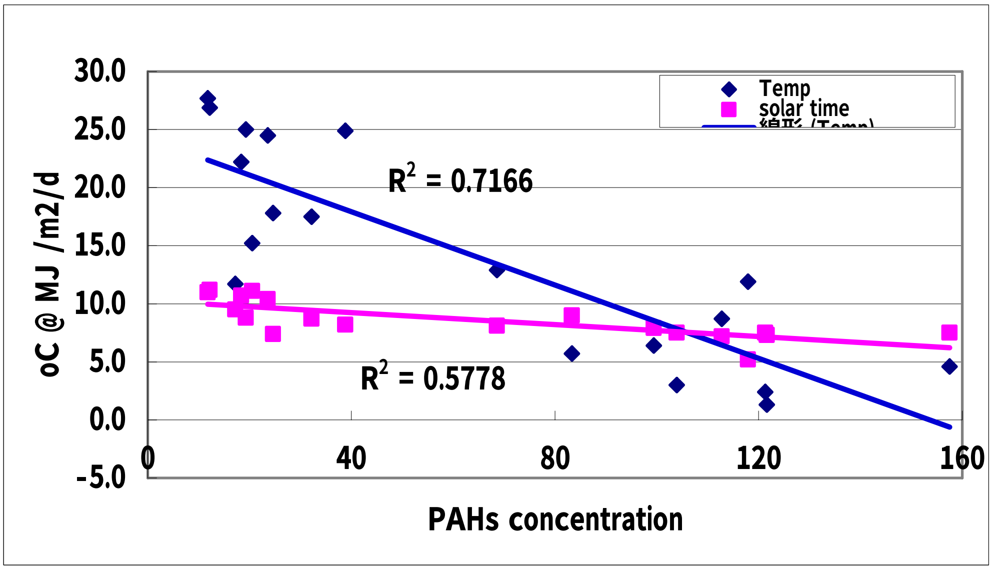
Principle Component Analysis (PCA) of Total concentration of aerosol PAHs

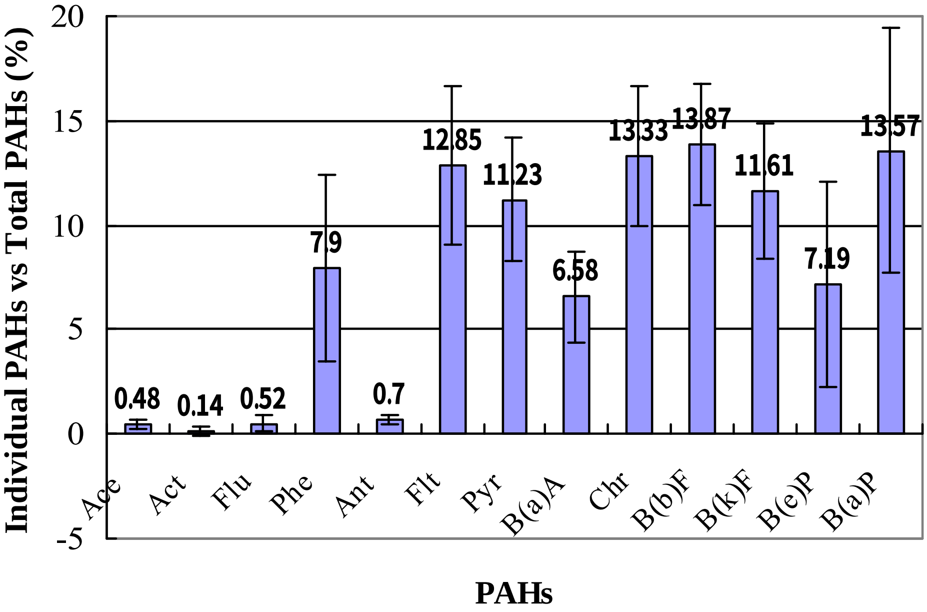
	Factor 1 (F1)	**Factor 2 (F2)**	**Factor 3 (F3)**
% of total variance	***69.3***	***13.5***	***8.3***
Ace	**0.914**		
Act		0.655	0.451
Flu	**0.770**		
Phe	**0.842**		
Ant	**0.973**		
Flt	**0.920**		
Pyr	**0.948**		
BaA	0.535	0.363	0.508
Chr	**0.955**		
BbF	**0.920**		
BkF	**0.866**	0.367	
BeP	**0.784**	0.355	
BaP	**0.840**	0.356	0.327

those with values greater than 0.7 are in bold


3 rings PAHs
4 rings PAHs
5 rings PAHs
PAHs concentration (ug/g aerosol)
160.0
140.0
120.0
100.0
80.0
60.0
40.0
20.0
0.0
2000/6/9
2000/6/16
2000/7/14
2000/8/11
2000/8/18
2000/8/25
2000/10/6
2000/11/10
2000/11/17
2000/11/24
2000/12/1
2000/12/8
2000/12/15
2000/12/22
2001/1/12
2001/2/19
2001/3/23
2001/5/11
2001/5/26
Sampling date


60
50
40
30
20
10
0
Frequency(%)
<0.1
0.1-0.2
0.3-0.5
0.6-0.8
0.9-1.0
Benzo(a)pyrene concentration range (ng/m3)


Temp
solar time
線形 (Temp)
R² = 0.7166
R² = 0.5778
oC @ MJ /m2/d
30.0
25.0
20.0
15.0
10.0
5.0
0.0
-5.0
0
40
80
120
160
PAHs concentration


Individual PAHs vs Total PAHs (%)
20
15
10
5
0
-5
0.48
0.14
0.52
7.9
0.7
12.85
11.23
6.58
13.33
13.87
11.61
7.19
13.57
Ace
Act
Flu
Phe
Ant
Flt
Pyr
B(a)A
Chr
B(b)F
B(k)F
B(e)P
B(a)P
PAHs