



Comparison of polycyclic aromatic hydrocarbon pollution in Chinese and Japanese residential air

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Abstract

Comparative studies on polycyclic aromatic hydrocarbon (PAH) pollution in residential air of Hangzhou (China) and Shizuoka (Japan) were conducted in summer (August, 2006) and winter (January, 2007). Total concentrations of 8 PAHs ranged from 7.1 to 320 ng/m³ and 0.15 to 35 ng/m³ in residential air of Hangzhou and Shizuoka, respectively. Air PAH concentrations in smoking houses were higher than that in nonsmoking houses. In nonsmoking houses, mothball emission and cooking practice were the emission sources of 2- and 3-ring PAHs in Hangzhou, respectively. The 2- and 3-ring PAHs were from use of insect repellent, kerosene heating and outdoor environment in nonsmoking houses in Shizuoka. The 5- and 6-ring PAHs in residential air were mainly from outdoor environment in both cities. Toxicity potencies of PAHs in residential air of Hangzhou were much higher than that in Shizuoka.

Key words: polycyclic aromatic hydrocarbons; residential air; emission source

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Introduction

Humans spend most of their lives in indoor environments, so the quality of indoor air has significant impact on human health. Assessment of human health risks from exposure of air pollution requires a detail understanding of pollutant levels in indoor environments.

Polycyclic aromatic hydrocarbons (PAHs) are a group of widespread environmental pollutants that consist of two or more fused benzene rings, and arise from a variety of combustion processes (Li et al., 2005). The predominant PAH sources to the atmosphere include motor vehicles, power generation, incineration and biomass burning (Mastral and Callen, 2000; Korenaga et al., 2001; Naumova et al., 2002; Ohura et al., 2004a). In indoor environments, PAHs are generated from cooking, smoking, incense burning and so on (Liu et al., 2001; Lung and Hu, 2003; Lu and Zhu, 2007).

The first survey of air PAH pollution was carried out in London in the 1950s (Waller, 1952), and since then, routine monitoring have been conducted at several locations around the world (Menichini, 1992; Harrison et al., 1996; Gustafson and Dickhut, 1997; Dimashki et al., 2001; Prevedouros et al., 2004). Most of these studies focused on PAH pollution in outdoor air. Since people nowadays

spend most of their time indoor, PAH pollution in indoor air has received increasing attention (Mitra and Ray, 1995; Li and Ro, 2000; Ohura et al., 2004b; Menichini et al., 2007). Several studies showed that indoor air PAH concentrations were higher than outdoor (Chuang et al., 1991; Liu et al., 2001; Lu et al., 2008). In addition, PAHs were found to be mostly associated with fine particles with diameters less than 2.5 μm (Koyano et al., 2001; Kume et al., 2007), which can reach the lower respiratory tract in human body, thus raising the concerns about their effects on human health.

Indoor air PAHs, especially low molecular weight ones, mainly come from indoor emission sources. Different life styles can result in different pollution patterns of PAHs in residential air. For example, using of mothball would largely emit naphthalene (Liu et al., 2001), whereas cooking fume is abundant in 3-ring PAHs (Zhu and Wang, 2003). However, little information on the effect of life styles on residential air PAH pollution is available. Resident life styles such as cooking practice in Chinese and Japanese are different. Chinese usually fry and broil food in residences, however, Japanese used to boil food or eat it freshly. Therefore, Hangzhou in China and Shizuoka in Japan were selected to study the effect of resident life styles on indoor air PAH pollution.

Hangzhou (30°15'N, 120°10'E), which has about 4.1

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million inhabitants, is the capital city of Zhejiang Province in China. The annual average rainfall and temperature are 1456 mm and 17.8°C, respectively. Shizuoka (34°59'N, 138°23'E) is the capital city of Shizuoka prefecture in Japan, and has about 0.7 million inhabitants. The annual average rainfall and temperature of Shizuoka are 2322 mm and 16.3°C, respectively. In both two cities, automobile exhaust is the main source of PAHs in ambient air (Ohura et al., 2004a; Zhu and Wang, 2005).

In the present study, PAH pollution levels in residential air were measured in Hangzhou and Shizuoka. The objectives of this study were: (1) to assess the PAH concentrations in indoor and outdoor air in selected residences; (2) to assess the influence of resident life styles on indoor PAH pollution, and clarify the emission sources of PAHs in residential air of the two cities.

1 Materials and methods

1.1 Sampling sites

The monitoring program in Hangzhou was carried out during the summer (August, 2006) and the winter (January, 2007). A total of 26 houses (6 houses in both seasons) were selected. In each house, samples were collected from main living area of the house, and outdoor samples were collected simultaneously in balcony. All samples were collected for 12 hr.

The monitoring program in Shizuoka was carried out during the same sampling periods. A total of 45 houses (21 houses in both seasons) were selected. In each house, samples were collected from main living area of the house, and outdoor samples were collected simultaneously in balcony. Air samples were collected for 24 hr because of relative low PAH concentrations in Shizuoka.

1.2 Air sampling and analysis

The procedures of air sampling and analysis were described in our previous publication (Lu and Zhu, 2007). In general, vapor PAHs were adsorbed using XAD-2 (Supelco, USA), which was cleaned with dichloromethane and methanol until peak of PAHs could not be found in HPLC. Particulate PAHs were collected with 25 mm glass fiber filters (GF, Whatman, England), which were preheated at 400°C for 6 hr to remove organic compounds before sampling. Both XAD-2 tubes and filters were connected with a mini-pump (Xingyu, China for Chinese samples; Sibata Scientific Technology, Japan for Japanese samples) for air sampling. The sampling flow rate was 1.0 L/min.

After air sampling, the XAD-2 were poured into a 25-mL glass stoppered tube containing 20 mL mixture of dichloromethane and acetonitrile ($V/V = 3/2$). The glass fiber filters were cut into pieces, placed in a 25-mL glass stoppered tube containing 10 mL dichloromethane. The samples were sonicated for 30 min while the water in the ultrasonic bath was replaced frequently to prevent overheating. Subsequently, 10 mL extracts of XAD-2 and 5 mL extracts of glass fiber filters were transferred into new glass stoppered tubes. Extracts with 30 μ L dimethyl

sulfoxide (DMSO) were evaporated under a gentle flow of nitrogen gas at room temperature and then 970 μ L acetonitrile was added.

The samples from Hangzhou sites were analyzed for the following 16 PAHs: naphthalene (NA), acenaphthylene (ACY), acenaphthene (AC), fluorene (FLUOR), phenanthrene (PHEN), anthracene (AN), fluoranthene (FLUR), pyrene (PY), benzo[a]anthracene (BaA), chrysene (CHRY), benzo[b]fluoranthrene (BbF), benzo[k]fluoranthrene (BkF), benzo[a]pyrene (BaP), dibenzo[a,h]anthracene (DA), benzo[ghi]perylene (BP), indeno[1,2,3-cd]pyrene (IN). The air samples from Shizuoka sites were analyzed for the following 8 PAHs: BaA, CHRY, BbF, BkF, BaP, DA, BP, IN. PAHs sampled in residential air of Hangzhou were determined by HPLC (Agilent, USA) containing a Lichrospher PAH column (250 mm \times 4.6 mm, Agilent, USA), a fluorescence detector and an ultraviolet detector (determine ACY only). PAHs sampled in residential air of Shizuoka were determined by HPLC (Hitachi, Japan) containing a Wakosil-II PAH column (250 mm \times 4.6 mm, Wako Chemicals, Japan) and a fluorescence detector (Hitachi, Japan). Thus, only 8 PAHs (BaA, CHRY, BbF, BkF, BaP, DA, BP, IN) simultaneously determined in both cities were compared to discuss the difference of residential air PAH pollutions in the two cities.

1.3 Quality control

A strict regime of quality control was operated in the experiment. Before the sampling program, PAH recovery studies were undertaken to demonstrate the reliability of the method. Working standard solution was made up from PAH stock solution purchased from Supelco (USA). Five pieces of separate XAD-2 and glass fiber filters were spiked with the working solution including the determined PAHs, then extracted and analyzed in the same way as the samples. The recoveries of each PAH were measured for vapor phase and particulate phase. For the samples collected in Hangzhou, the recoveries of 16 PAHs for vapor phase and particulate phase ranged from (85.3 \pm 4.9)% to (98.7 \pm 3.8)% except for NA which was (77.0 \pm 2.7)% and (78.8 \pm 5.4)%, respectively, and the relative standard deviations of the recoveries of 16 PAHs were less than 5.5%. For the samples collected in Shizuoka, the recoveries of 8 PAHs for two phases ranged from (96.4 \pm 1.8)% to (102.4 \pm 4.6)%, and the relative standard deviations of the recoveries were less than 8.8%.

2 Results and discussion

2.1 PAH concentrations in residential air

PAH concentrations in residential air of two cities are presented in Table 1. In summer, total concentrations of 8 PAHs (Σ PAHs) ranged from 7.1 to 320 ng/m³ and 0.15 to 32 ng/m³, with an average of 47 ng/m³ and 5.2 ng/m³ in residential air of Hangzhou and Shizuoka, respectively. In winter, the corresponding concentrations varied from 8.1 to 160 ng/m³ and 0.26 to 35 ng/m³, with an average

Table 1 PAH concentrations in summer and winter

PAHs	Hangzhou in summer			Shizuoka in summer			Hangzhou in winter			Shizuoka in winter		
	Mean	Max	Min	Mean	Max	Min	Mean	Max	Min	Mean	Max	Min
BaA (ng/m ³)	35	2.6E2	4.4	0.22	1.9	0.011	12	40	4.2	0.38	1.9	0.016
CHRY (ng/m ³)	7.3	35	2.2	0.70	5.0	0.032	6.1	13	1.7	0.86	5.3	0.029
BbF (ng/m ³)	1.4	8.3	0.17	1.2	5.1	0.031	5.6	14	0.57	1.4	8.9	0.055
BkF (ng/m ³)	0.69	3.7	0.072	0.47	3.0	0.010	2.4	59	0.32	0.44	2.5	0.019
BaP (ng/m ³)	0.61	2.1	0.11	0.65	5.0	0.017	4.4	12	0.52	0.85	4.7	0.039
DA (ng/m ³)	0.33	2.1	nd	0.13	0.74	nd	0.69	3.2	0.18	0.14	0.70	nd
BP (ng/m ³)	1.2	3.6	0.15	0.94	5.5	0.026	4.3	9.4	0.58	1.1	5.8	0.053
IN (ng/m ³)	0.65	2.1	nd	0.90	5.4	0.018	4.9	13	nd	0.96	5.1	0.039
∑PAHs (ng/m ³)	47	3.2E2	7.1	5.2	32	0.15	40	1.6E2	8.1	6.1	35	0.26

nd: not detected.

of 40 and 6.1 ng/m³ in Hangzhou and Shizuoka, respectively. The results indicated that ∑PAHs in Hangzhou were significantly higher than that in Shizuoka. Among the 8 PAHs, concentrations of 4-ring PAHs (BaA and CHRY) were much higher in residential air of Hangzhou, whereas concentrations of 5- to 6-ring PAHs were similar in residential air of two cities.

BaA was observed to be the most abundant in residential air of Hangzhou, which could be emitted from cooking practice (Zhu and Wang, 2003). The concentrations of BaA accounted for 62.0% to 80.0% and 24.4% to 52.0% of ∑PAHs in summer and winter, respectively. In residential air of Shizuoka, BbF was the most abundant, accounting for 16.2% to 20.5% and 21.3% to 25.6% of ∑PAHs in summer and winter, respectively. Most of high molecular weight (MW) PAHs is mutagenic and carcinogenic to humans (Caricchia et al., 1999; Maliszewska-Kordybach, 1999). BaP concentrations were observed ranging from 0.11 to 12 ng/m³ and 0.017 to 5.0 ng/m³ in residential air of Hangzhou and Shizuoka, respectively, which indicated that the concentrations were much higher than those in Houston (0.0027–1.1 ng/m³) and Elizabeth (0.0055–0.23 ng/m³) (Naumova et al., 2002).

The average summer/winter ratios of PAH concentrations were calculated to assess the seasonal variation of individual PAHs (Fig. 1). Concentrations of 4-ring PAHs were higher in summer than those in winter in Hangzhou, with corresponding summer/winter ratios higher than 1.0.

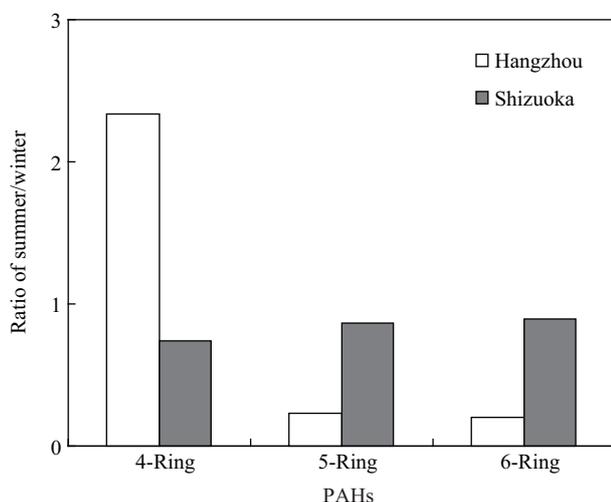


Fig. 1 Average summer/winter ratios of PAH concentrations.

However, the opposite case was found in Shizuoka. In addition, the ratios for 5- to 6-ring PAHs were both lower than 1.0, especially in Hangzhou. This phenomenon may be attributed to photodegradation to related compounds in summer, and reductions in combustion temperature (Freeman and Cattell, 1990; Harrison et al., 1996; Gustafson and Dickhut, 1997; Papageorgoulou et al., 1999).

2.2 Relationship between indoor and outdoor PAH concentrations

The average ratios of indoor/outdoor PAH concentrations are given in Fig. 2. In general, the variation trend of indoor/outdoor ratios was similar in the two cities. The ratios of 4-ring PAHs were higher than 1.0 in summer in residential air of Hangzhou, which indicated that there were indoor emission sources for 4-ring PAHs. However, the ratios of 5- and 6-ring PAHs were lower than 1.0, especially in winter, indicating that they mainly come from outdoor emission sources.

Significant correlations between the concentrations of 5- and 6-ring PAHs in indoor and outdoor air were observed in both cities ($r \geq 0.500$, $p < 0.05$) (Table 2). The correlations for those in Hangzhou were more significant in winter than in summer, however, opposite case was observed in Shizuoka. The results strongly supported the above views that concentrations of 5- to 6-PAHs were predominated by outdoor sources, especially during the winter in Hangzhou and during summer in Shizuoka.

2.3 Emission sources of PAHs in residential air

In this section, some data shown in our previous publications (Ohura et al., 2004b; Zhu et al., 2009) were used to assess the influence of resident life styles on PAH pollution in residential air comprehensively.

Smoking is an important source of PAHs in residential air (Gundel et al., 1995; Liu et al., 2001). However, there were not enough houses smoked during sampling period in the two cities, so the influence of smoking

Table 2 Relationship between indoor and outdoor PAH concentrations

PAHs	Hangzhou		Shizuoka	
	Summer	Winter	Summer	Winter
4-Ring	0.389	0.497*	0.425*	0.258
5-Ring	0.784**	0.843**	0.895**	0.565**
6-Ring	0.767**	0.896**	0.766**	0.500*

Significance at * $p < 0.05$; ** $p < 0.01$.

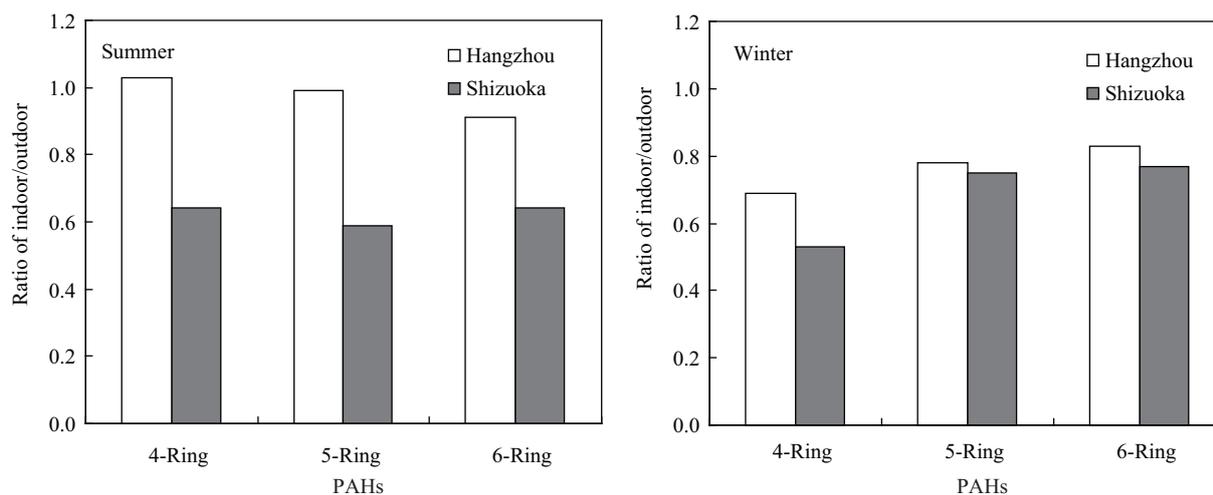


Fig. 2 Average indoor/outdoor ratios of PAH concentrations.

Table 3 Correlation coefficient for emission sources and PAH concentrations in nonsmoking residential air of Shizuoka

	2-Ring	3-Ring		4-Ring	5-Ring	6-Ring
		Summer	Winter			
Insect repellent	0.108*					
Kerosene heating			0.653**			
Outdoor environment		0.743**		0.820**	0.760**	0.745**
House age					-0.332**	-0.360**

Significance at * $p < 0.05$; ** $p < 0.01$.

on PAH concentrations in residential air could not be analyzed statistically. Nevertheless, the differences of PAH concentrations still could be observed between the smoking and nonsmoking residences. In Hangzhou, average concentrations of BaP in summer were 0.84 ng/m^3 ($0.31\text{--}2.1 \text{ ng/m}^3$) and 0.60 ng/m^3 ($0.11\text{--}1.6 \text{ ng/m}^3$) in smoking and nonsmoking houses, respectively; in Shizuoka, the corresponding concentrations were 0.25 ng/m^3 ($0.16\text{--}1.2 \text{ ng/m}^3$) and 0.19 ng/m^3 ($0.048\text{--}0.84 \text{ ng/m}^3$) in smoking and nonsmoking houses, respectively. The results indicated that BaP concentrations in smoking houses were higher than those in nonsmoking houses.

Factor analysis was conducted to clarify emission sources of PAHs in nonsmoking residential air of Hangzhou. The rotated component matrix of PAH concentrations was calculated, and the results were given in our previous publication (Zhu et al., 2009). Four factors were selected, and the variances were 41.2%, 24.1%, 10.0% and 8.9% for factor 1 to 4, respectively. Factor 1 represented outdoor environment, because it had high loading on 5- to 6-ring PAHs and researchers indicated that high ring PAHs were usually emitted from outdoor sources, such as gasoline combustion engines, rather than from most indoor combustion processes such as cooking and smoking (Miguel et al., 1998). Factor 2 represented cooking practice, because it was highly loaded on 3-ring PAHs such as AC, FLUOR and PHEN. The fingerprint of fume from Chinese conventional cooking method was the abundance of 3-ring PAHs (Zhu and Wang, 2003). Factor 3 represented mothball emission because of its high loading on NA. Mothballs which were used to protect clothes were commonly used in selected houses of Hangzhou, and NA was largely emitted from mothballs

(Liu et al., 2001). There was no other emission source during sampling period in the houses, therefore factor 4 represented unknown source and were speculated as the environmental background concentrations of PAHs. However, further study should be conducted to clarify this factor. Therefore, mothball emission, cooking practice, outdoor environment and unknown source (may be environmental background concentrations of PAHs) were taken as the emission sources of PAHs in nonsmoking residential air of Hangzhou.

Multiple regression analysis based on indoor and outdoor PAH concentrations and the responses to questionnaire about resident life styles were conducted in nonsmoking residential air of Shizuoka (Table 3), and the results were shown in our former study (Ohura et al., 2004b). Concentrations of 2-ring PAHs (NA) were correlated well with the use of insect repellent, indicating that the use of insect repellent was the main source of NA in residential air in Shizuoka. 3-Ring PAHs emitted from two sources in summer and winter. In summer, they were mainly transported from outdoor air, whereas kerosene heating was the main emission source in winter. Concentrations of 4- to 6-ring PAHs were significantly affected by the corresponding outdoor concentrations, showing that these PAHs were mainly from outdoor environment. In addition, concentrations of 5- to 6-ring PAHs were also affected by the house age. Newly built houses may experience higher concentrations of these PAHs.

The above results showed that 2- to 3-ring PAHs in residential air were from different emission sources in the two cities. The use of mothball and insect repellent took the responsibility for NA in nonsmoking residential air of Hangzhou and Shizuoka, respectively. In Hangzhou,

people usually fry and broil food such as fish and pork in residences, which can emit 3-ring PAHs largely (Zhu and Wang, 2003). In Shizuoka, people used to boil food or eat it freshly, which can emit much less 3-ring PAHs, and these PAHs were mainly from outdoor environment and kerosene. In addition, 5- to 6-ring PAHs in the two cities were both predominated by outdoor environment.

2.4 Toxicity assessment of PAHs pollution in residential air

Toxicity equivalency factor (TEF) based on BaP could be used to assess the toxicity potencies of various PAHs in respect to inhalation cancer risks to humans (Nisbet and LaGoy, 1992). Average TEF-adjusted PAH concentrations (Σ TEF-PAHs) in residential air are shown in Table 4. The results indicated that average Σ TEF-PAHs in Hangzhou were 4.5 times and 5.8 times higher than those in Shizuoka in summer and winter, respectively. In Hangzhou, average Σ TEF-PAHs were mainly contributed from BaA (72.9%) in summer and BaP (57.2%) in winter. Average Σ TEF-PAHs were dominated by BaP (60.4% in summer and 64.0% in winter) in both seasons in Shizuoka.

Table 4 Average TEF-adjusted concentrations of PAHs in residential air (unit: ng/m³)

PAHs	TEF	Hangzhou		Shizuoka	
		Summer	Winter	Summer	Winter
BaA	0.1	3.5	1.2	0.022	0.038
CHRY	0.01	0.073	0.061	0.0070	0.0086
BbF	0.1	0.14	0.56	0.12	0.14
BkF	0.1	0.069	0.24	0.047	0.044
BaP	1	0.61	4.4	0.65	0.85
DA	1	0.33	0.69	0.13	0.14
BP	0.01	0.012	0.043	0.0094	0.011
IN	0.1	0.065	0.49	0.09	0.096
Σ TEF-PAHs		4.8	7.7	1.1	1.3

3 Conclusions

Total concentrations of 8 PAHs varied from 7.1 to 320 ng/m³ and 0.15 to 35 ng/m³ in residential air of Hangzhou and Shizuoka, respectively. Among these PAHs, BaA was observed to be the most abundant in Hangzhou owing to its large emission from cooking fume, whereas the most abundant one was BbF in Shizuoka. The ratios of indoor/outdoor PAH concentrations showed that 4-ring PAHs were influenced by indoor sources, whereas 5 to 6-ring PAHs were predominated by outdoor sources in both cities.

PAH concentrations in residential air of smoking houses were higher than those in nonsmoking houses. In nonsmoking houses, mothball emission, cooking practice, outdoor environment and unknown source (may be environmental background concentrations of PAHs) were the emission sources of PAHs in residential air of Hangzhou. However, PAHs concentrations were affected by use of insect repellent, kerosene heating, outdoor environment and house age in nonsmoking residential air of Shizuoka. Toxicity potencies of PAHs in residential air of Hangzhou

were much higher than those in Shizuoka. In Hangzhou, BaA and BaP made the largest contribution to Σ TEF-PAHs in summer and winter, respectively. In Shizuoka, the largest contributor in both seasons was BaP.

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