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Effect of central ventilation and air conditioner system on the concentration and health risk from airborne polycyclic aromatic hydrocarbons

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Abstract

Central ventilation and air conditioner systems are widely utilized nowadays in public places for air exchange and temperature control, which significantly influences the transfer of pollutants between indoors and outdoors. To study the effect of central ventilation and air conditioner systems on the concentration and health risk from airborne pollutants, a spatial and temporal survey was carried out using polycyclic aromatic hydrocarbons (PAHs) as agent pollutants. During the period when the central ventilation system operated without air conditioning (AC-off period), concentrations of 2–4 ring PAHs in the model supermarket were dominated by outdoor levels, due to the good linearity between indoor air and outdoor air ($r_p > 0.769$, $p < 0.05$), and the slopes (1.2–4.54) indicated that ventilating like the model supermarket increased the potential health risks from low molecular weight PAHs. During the period when the central ventilation and air conditioner systems were working simultaneously (AC-on period), although the total levels of PAHs were increased, the concentrations and percentage of the particulate PAHs indoors declined significantly. The BaP equivalency (BaP_{eq}) concentration indicated that utilization of air conditioning reduced the health risks from PAHs in the model supermarket.

Key words: particulate; gaseous; PAHs; health risk; public place
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Introduction

During the past two decades, indoor air pollution has drawn increasing attention all over the world. Many pollutants that people respire indoors come from outdoor sources, and long term exposure to them may cause cardiovascular disease, cancer, respiratory disease and so on (Calderon-Garciduenas et al., 2004; Knutsen et al., 2004; Schwartz, 1996; Schwarze et al., 2006). The exchange of pollutants between indoor and outdoor air is achieved by ventilation, therefore the ventilation pattern will have a significant effect on indoor air quality. In previous studies, the effects of open windows, ventilation frequency and air conditioner use on levels of indoor airborne pollutants were considered (Levy et al., 2002; Ohura et al., 2004). However, as far as we know there have been few studies concerning the influences of central ventilation systems on the concentrations and health risk from airborne pollutants.

Polycyclic aromatic hydrocarbons (PAHs), many of which are strongly potent carcinogens and/or mutagens (Alfheim et al., 1984), are generated from incomplete pyrolysis of organic matter such as fossil fuel, cigarettes and coal. Residential heating, power generation, industry and automobile exhaust produce the dominant mass of PAHs outdoors (Baek et al., 1991; Maliszewska-Kordybach, 1999; Mastral and Callén, 2000), especially for cities; while tobacco smoke, cooking and heating are the major indoor sources (Baek et al., 1991; Gundel et al., 1995; Maliszewska-Kordybach, 1999). As smoking is gradually being forbidden in public places in recent years in China, the PAHs that people are exposed to in public places are mainly from outdoor sources. Besides, PAHs exist both in the gaseous and particulate phase. The proportion of either phase is determined by the molecular structure of each PAH. Generally, the ratio of gaseous to particulate PAH decreases as the molecular weight increases (Pankow, 1987). Therefore, PAHs are ideal for studying the effect of ventilation patterns on the concentrations of both gaseous and particulate pollutants.

This comparative study on the indoor to outdoor ratios of PAH concentrations for buildings was conducted to evaluate the effects of the central ventilation and air conditioner systems on PAH concentrations, as well as potential health risks.

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1 Materials and methods

1.1 Sampling sites

Two large supermarkets (SMs) and two shopping centers (SCs), with identical central ventilation and air conditioner systems, were carefully selected in Hangzhou, China. The central air conditioner systems operated commonly during May to September to keep indoor temperature below outdoor temperature (AC-on period), while indoor temperature is higher than outdoor temperature when the air conditioner is not working (AC-off). Central ventilation is the only pattern for the air exchange between indoor and outdoor air in both AC-on and AC-off periods. The largest supermarket in Hangzhou, with business area of 24,000 m^2, was chosen as a model microenvironment with a central ventilation and air conditioner system for long-term air sampling. It was 50 meters away from a main street, around which automobile exhaust was the dominant source of airborne PAHs. The ventilation rate of the model supermarket (MS) was about 450 m^3/min. Twelve non-smoking residences around the MS with natural ventilation were selected as the control. Other details of SMs, SCs and residences were presented in our previous study (Lu et al., 2008).

1.2 Air sampling

Gaseous and particulate PAHs were collected by 2.5 g XAD-2 adsorbent (Supelco, USA) and 25 mm glass fiber filter (GFF, Whatman, England) respectively. A mini-pump (DDY-1.5, Xingyu, China), which was placed at a height of 1.5–1.8 m above the ground, was used for air sampling at a flow rate of 1.0 L/min for 8 hr (8:30–16:30). The flow rates were measured before and after the sampling, with deviations less than 2%. Windows were closed during sampling in residences. Six samplers were set in the region of household appliances, cosmetics and daily necessaries respectively, and two samplers were set on the roof of the SMs and SCs.

Fourteen samples in SMs and SCs were collected during March to April, 2006. A total of 58 samples for residences were collected in January, 2007. A total of 61 samples in the MS were taken during 2007 to 2010, of which 26 samples were taken during AC-off periods.

1.3 Analysis of PAHs samples

The determination of PAHs was described in our previous paper (Liu et al., 2001). In general, after air sampling, XAD-2 adsorbent samples were poured into a stoppered 25 mL glass tube with a 20 mL mixture of dichloromethane and acetonitrile (V/V = 3/2), while glass fiber filters were cut into pieces and placed in a stoppered 25 mL glass tube with 10 mL dichloromethane (Supelco, USA). Subsequently, the samples were sonicated for 30 min. During the sonication, the water in the ultrasonic bath was replaced frequently to prevent overheating. Then 10 mL extracts of XAD-2 and 5 mL extracts of glass fiber filters were transferred into other glass stoppered tubes. Extracts with 30 μL dimethyl sulfoxide (Supelco, USA) were evaporated under a gentle flow of nitrogen gas at room temperature and then 970 μL acetonitrile was added (Supelco, USA).

The following 14 PAHs (Supelco, USA) were determined in the air samples: naphthalene (NA), acenaphthene (AC), 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), and indeno(1,2,3-cd)pyrene (IN). PAHs were determined using UPLC (ACQUITY, Waters, USA) equipped with a C18 column (ACQUITY UPLC 1.7 μm, 2.1 × 150 mm, Waters, USA) and a FLR detector. Data were analyzed by SPSS statistics 17.0.

1.4 Quality control

XAD-2 adsorbent was cleaned via soxhlet extraction in dichloromethane and methanol until no signal of PAHs was detected in UPLC. GFFs were heated at 400°C for 6 hr prior to use. PAHs corresponding to 100–800 ng/m^3 in vapor phase and 2–10 ng/m^3 in particulate phase were spiked onto XAD-2 and GFFs respectively to undertake a PAHs recovery study (n = 5). The recovery of NA was 77% for vapor phase and 78% for particulate phase. The recoveries of the other 13 PAHs all exceeded 85%, and the relative standard deviations of the recoveries of the 14 PAHs were less than 3.8%.

2 Results and discussion

2.1 Higher potential risks from the PAHs in SMs & SCs than in residences

A comparative study of SMs & SCs and residences, using central ventilation and natural ventilation respectively, based on our previous work, was carried out in the AC-off period (Lu et al., 2008; Zhu et al., 2009). **Table 1** shows that the average indoor to outdoor ratio (I/O) of 2–4 ring PAHs for SMs & SCs is 3.98 (0.94–12.51), significantly higher than 1.03 (0.63–1.68) for residences ($p < 0.05$), while the I/O of 5–6 ring PAHs are similar between SMs & SCs and residences ($p < 0.05$), except for BP ($0.01 < P < 0.05$). This indicated that people were faced with a higher health risk from 2–4 ring PAHs in SMs & SCs than in residences.

Two facts could cause the diversity of I/O for 2–4 ring PAHs. Firstly, there may be some stronger sources existing in SMs & SCs, for the I/O is always greater than 1 when significant sources were found indoors (Halsall et al., 2008; Liu et al., 2001; Ohura et al, 2004). Secondly, the different ventilation patterns used in the two kinds of microenvironment may greatly influence the outdoor-indoor air exchange.

Table 1 Indoor to outdoor ratios (I/O) of PAHs in SMs & SCs and residences

PAHs	Ring	Mean I/O		*p* value
		SMs & SCs	Residences	
NA	2	2.76	1.68	0.01–0.05
AC	3	12.51	1.57	< 0.01
PHEN	3	2.53	1.29	0.01–0.05
AN	3	9.30	0.95	< 0.01
FLUR	4	1.19	0.64	< 0.01
PY	4	1.31	0.78	< 0.01
BaA	4	1.65	0.69	< 0.01
CHRY	4	0.94	0.63	0.01–0.05
BbF	5	0.87	0.70	> 0.05
BkF	5	0.85	0.72	> 0.05
BaP	5	0.89	0.81	> 0.05
DA	5	1.09	0.80	> 0.05
BP	6	1.25	0.82	0.01–0.05
IN	6	0.96	0.79	> 0.05

Some data in **Table 1** have been published in our previous works (Lu and Zhu, 2008, 2009).
SMs: supermarkets; SCs: shopping centers.

2.2 Source identification of indoor PAHs

To evaluate the influence of large amounts of commodities with similar function placed together in a certain region, samplers were set at three different regions in SMs and SCs such as the household appliance region, cosmetic region and daily necessaries region. As shown in **Fig. 1**, when the outdoor concentrations of naphthalene (NA) are below 420.58 ng/m^3, the indoor concentrations in different regions exhibit a certain order, cosmetic region > daily necessaries region > household appliance region, which indicated that certain sources of NA existed in the cosmetic and daily necessaries regions. When the outdoor concentration of NA was over 420.58 ng/m^3, there was no notable difference found among these regions, largely owing to the effect of outdoor NA. No significant sources of other PAHs were found in the SMs and SCs. Therefore, the relatively higher I/O of 2–4 ring PAHs in SMs and SCs compared to that in residences discussed in Section 2.1 was largely caused by different ventilation patterns.

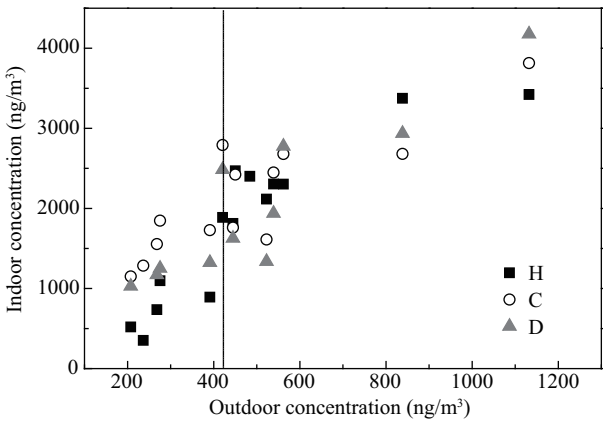

Fig. 1 Correlation between the indoor and outdoor concentrations of naphthalene NA. H: household appliance region; C: cosmetic region; D: daily necessaries region.

Table 2 Linear correlations of PAHs between indoors and outdoors

PAHs	AC-off			AC-on		
	n	r_p	Slope	*n*	r_p	Slope
NA	9	0.989**	3.18	7	0.953**	5.86
AC	7	0.950**	4.54	7	0.157	–
PHEN	9	0.970**	2.30	6	0.527	–
AN	9	0.940**	2.14	6	0.731	–
FLUR	8	0.874**	1.54	7	0.561	–
PY	8	0.954**	2.02	7	0.762*	0.46
BaA	7	0.725	–	7	0.725	–
CHRY	9	0.769*	1.20	7	0.827*	0.37
BbF	8	0.623	–	7	0.446	–
BkF	9	0.100	–	6	0.778	–
BaP	8	–0.507	–	7	0.215	–
DA	8	0.401	–	6	0.412	–
BP	8	0.625	–	6	0.184	–
IN	–	–	–	5	0.080	–

** $P < 0.01$; * $P < 0.05$.

2.3 Effect of central ventilation on PAH transfer

We analyzed the data obtained in the model supermarket (MS) by linear regression. **Table 2** shows that the PAHs could be divided into two parts during the AC-off period. First, the r_p^2 of 2–4 ring PAHs were greater than 0.526 (except for BaA, $p > 0.05$), among which NA had the best linearity ($r_p^2 = 0.978$, $p < 0.01$). This proved that the concentrations of indoor 2–4 ring PAHs in the MS were largely controlled by outdoor levels when central ventilation worked alone, and the higher I/O for SMs and SCs (shown in **Table 1**) was largely due to the ventilation pattern rather than the indoor sources; Second, the r_p^2 of 5–6 ring PAHs were below 0.388, showing poor linearity between indoor and outdoor concentrations ($p > 0.05$). To better understand this diversity, the phase distribution of outdoor PAHs and r_p^2 are both given in **Fig. 2**. As can be seen in this figure, the r_p^2 decreased significantly with the proportion of vapor phase, revealing a significant effect of the ventilation pattern on the particulate PAHs transfer. This was owing to the fact that the filters in the central ventilation system blocked a portion of the particles during the air transfer process from outdoor to indoor. As no notable reduction of the concentrations was observed for 5–6 ring PAHs in indoor air, there should be other factors influencing the levels of the indoor particulate PAHs, such as dust resuspension (Saraga et al., 2010).

The slopes of 2–4 ring PAHs in the MS were 1.20–4.54, which were much higher than those in the residences. It was more likely that the MS acted as a sorbent, which concentrated the 2–4 ring PAHs from outdoors. Therefore people in public places ventilating like the MS may face higher risks from 2–4 ring PAHs than in residences with natural ventilation.

Source identification of PAHs in MS was evaluated by factor analysis (calculating by SPSS Statistics 17.0). **Figure 3** shows that Factor 1 explained 65.53% and 77.05% of the variance of outdoor PAHs in the periods of AC-off

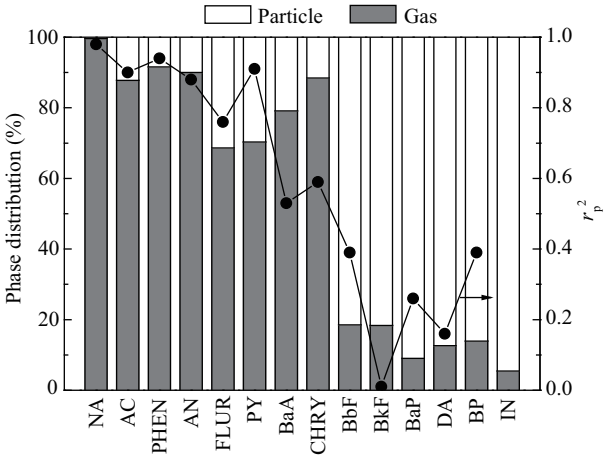

Fig. 2 Phase distribution and r_p^2 of PAHs in MS.

and AC-on, respectively. PAHs with high molecular weight (molecular weight) were mainly generated from high temperature splitting of organic matter (automobile exhaust). Factor 1 had high factor load on the high molecular weight PAHs and FLUR (Larsen and Baker, 2003; Li and Kamens, 1993; Simcik et al., 1999), which indicated that automobile exhaust was the major source of PAHs in the ambient air of the MS. The percentage variance of Factor 1 in the AC-off period was lower than that in the AC-on period, largely owing to the affects of coal burning in the AC-off period and climate change. The factors with high load on low molecular weight PAHs in indoor air were different from those outdoors, which was largely due to the fact that the filters in the central ventilation system blocked a portion of particles from outdoors, which thereby changed the relationship between the concentrations of the low and high molecular weight PAHs.

2.4 Effect of air conditioner on indoor PAHs levels

Table 2 shows that no linear correlation between PAHs indoors and outdoors was observed during the AC-on period, except for NA (r_p = 0.953, $p < 0.01$), PY (r_p = 0.762, $p < 0.05$) and CHRY (r_p = 0.827, $p < 0.05$), which was significantly different from the AC-off period. This is largely due to the fact that the air exchange between the indoor and outdoor air is lower when the air conditioner is operating than that when the central ventilation works alone. Moreover, the lower temperature indoors compared to outdoors would reduce the air exchange between indoor air and outdoor air.

As can be seen in **Table 3**, although the total concentration of PAHs in the AC-on period was slightly higher than that in AC-off period, the percentage of particulate PAHs in the AC-on period was much lower. **Figure 4** shows that the concentrations of particulate 4–6 ring PAHs outdoors were similar between the AC-off and AC-on periods, while those indoors in the AC-on period were significantly lower than in the AC-off period. This was largely due to the fact that the air conditioner system, which circulates the indoor air

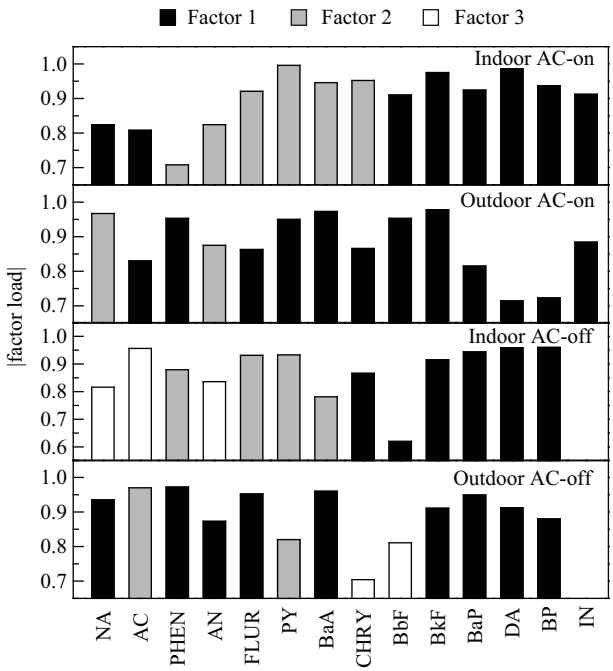

Fig. 3 Factor analysis of PAHs in MS.

through the ventilation ducts, thereby blocks a portion of particulate PAHs. The levels of gaseous PAHs in the AC-off period were higher than in the AC-on period. However, their difference indoors was smaller than that outdoors, mainly owing to the lower temperature indoors and the reduction of particulate PAHs by the air conditioner.

2.5 Assessment of health risk

Several approaches have been developed to evaluate the potencies of a complex mixture of PAHs with respect to inhalation cancer risks to humans. The BaP equivalency (BaP_{eq}) concentration is widely applied to assess toxicity potencies by using a list of toxic equivalency factors (TEFs) based on BaP (Petry et al., 1996). **Figure 5** shows that the indoor BaP_{eq} concentration was 2.14 ± 1.09 fold and 1.30 ± 0.65 fold higher than the outdoor BaP_{eq} concentration in the AC-off and AC-on periods, respectively. Although the concentration of the outdoor BaP_{eq} in the AC-on period was higher than that in the AC-off period, the indoor BaP_{eq} concentration in the AC-on period was 28.9% lower. This was due to the air conditioner reducing the levels of particulate PAHs, which have higher health risks.

Nowadays, central ventilation systems have been widely adopted in millions of public places in China. Based on the data we obtained, ventilation patterns should be selected carefully and designed properly to decrease the magnification effect of pollutants from outdoor sources by buildings. Although application of air conditioning along with central ventilation systems decreases the air exchange rate, it may decrease the health risks from outdoor pollutants, especially for particulate phases.

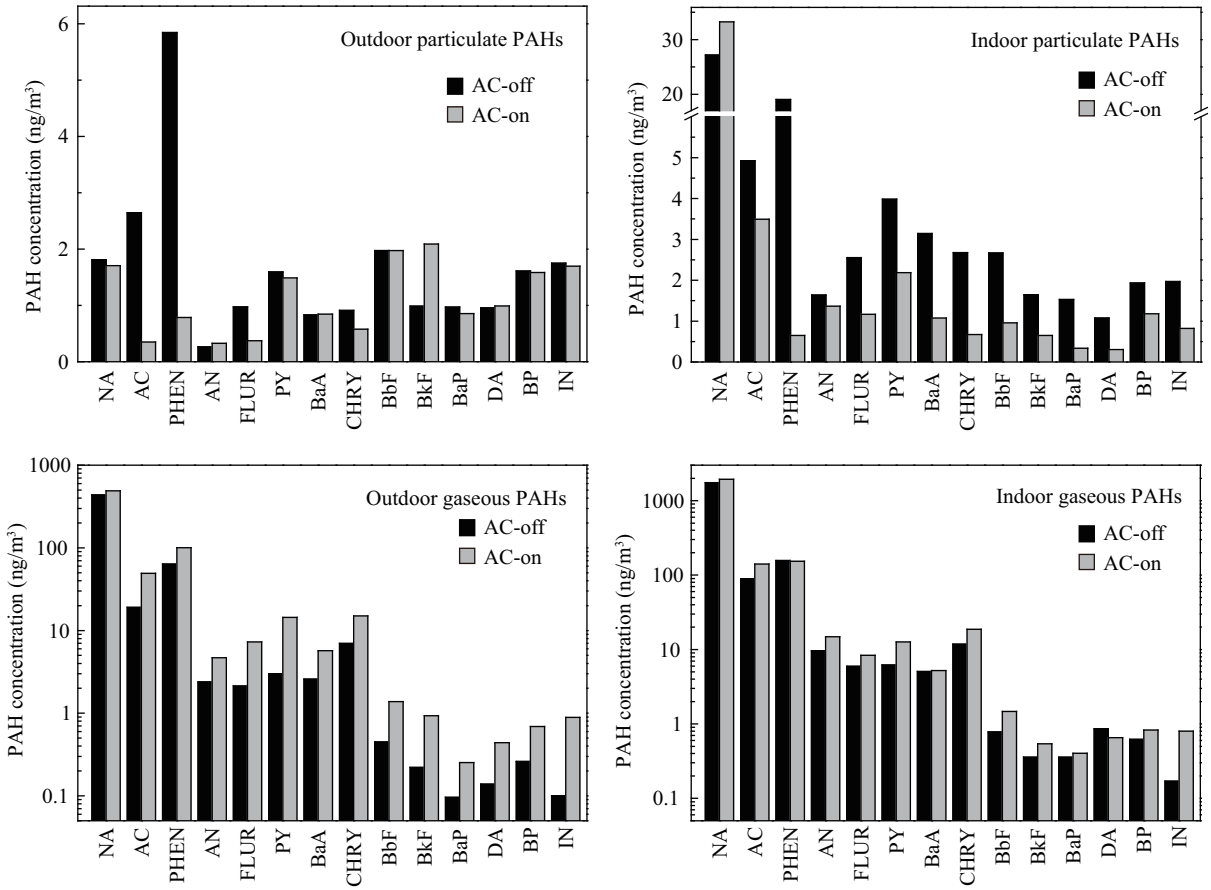

Fig. 4 Concentrations of gaseous and particulate PAHs in indoor and outdoor air.

Table 3 Average concentrations and phase distribution of PAHs in MS

PAHs	AC-off			AC-on		
	Concentration (ng/m^3)	Particulate (%)	Gaseous (%)	Concentration (ng/m^3)	Particulate (%)	Gaseous (%)
NA	1772.08	1.53	98.47	1968.78	1.69	98.31
AC	94.46	5.22	94.78	144.46	2.42	97.58
PHEN	176.13	10.81	89.19	154.33	0.42	99.58
AN	11.27	14.54	85.46	16.25	8.41	91.59
FLUR	8.51	30.00	70.00	9.50	12.26	87.74
PY	10.17	39.16	60.84	14.84	14.73	85.27
BaA	8.22	38.23	61.77	6.30	17.08	82.92
CHRY	14.46	18.51	81.49	19.34	3.48	96.52
BbF	3.45	77.38	22.62	2.43	39.42	60.58
BkF	2.00	82.18	17.82	1.19	54.39	45.61
BaP	1.89	81.08	18.92	0.74	45.32	54.68
DA	1.94	55.49	44.51	0.96	31.84	68.16
BP	2.55	75.66	24.34	2.01	58.81	41.19
IN	2.14	92.06	7.94	1.62	50.75	49.25
Total	2109.27	3.60	96.40	2342.72	2.05	97.95

3 Conclusions

The I/O ratios of the 2–4 ring PAHs for the shopping centers and supermarkets with central ventilation systems were significantly higher than those for residences with a natural ventilation pattern. In the model supermarket, a good linearity ($r_p > 0.874$, $p < 0.01$) was observed between low molecular weight PAHs in indoor and outdoor air in the AC-off period, which indicated that the concentrations of low molecular weight PAHs indoors were largely dominated by the outdoor levels when central ventilation worked alone. The slopes of the 2–4 ring PAHs in the MS were much higher than 1, revealing a magnification effect of the 2–4 ring PAHs levels outdoors. When central ventilation and air conditioning operated simultaneously, the concentrations of PAHs increased, while the levels

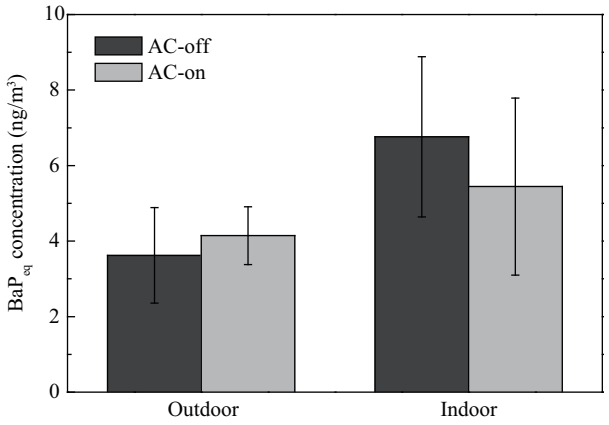

Fig. 5 BaP_{eq} concentrations of PAHs in the AC-on and AC-off period.

and percentages of indoor particulate PAHs decreased significantly. The I/O ratio of BaP_{eq} in the AC-off period was 1.65 times higher than that in the AC-on period, which shows that the operation of the air conditioner in the MS reduced the potential health risks of airborne PAHs.

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