

Pollution patterns of polycyclic aromatic hydrocarbons in tobacco smoke

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Abstract

Concentrations of polycyclic aromatic hydrocarbons (PAHs) in tobacco smoke of 12 commercial brand cigarettes were determined in a simulated chamber of 20.25 m³ in size. The total concentrations of 17 PAHs (\sum PAHs) in the chamber were 3500 and 1152 ng/m³ in vapor phase and particulate phase, respectively. In vapor phase, the yield of naphthalene (NA) appeared to be the most abundant (2462 ng/cig) followed by fluorene (FLUOR) and acenaphthylene (ACY), while the yield of benzo[ghi]perylene (BP) was the most abundant (259.7 ng/cig) in particulate phase followed by phenanthrene (PHEN) and FLUOR. The proportion of PAHs in particulate phase increased with increasing molecular weight. PAHs with two to six rings accounted for 40.2%, 35.3%, 11.7%, 7.6%, 5.2% of \sum PAHs, respectively. There was no obvious correlation between PAHs, benzo[a]pyrene (BaP) concentrations in tobacco smoke and smoking tar contents, nicotine contents. With the source fingerprint of PAHs in tobacco smoke, NA could be regarded as the marker of tobacco smoke source because of its largest contribution to \sum PAHs (40.2%), followed by FLUOR (12.7%) and ACY (9.8%). Further study indicated that more than 80% of BaP in indoor air of resident homes in Hangzhou was from tobacco smoke.

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1. Introduction

Polycyclic aromatic hydrocarbons (PAHs) are a well-known group of environmental pollutants and subject of intensive investigations. They are carcinogenic to human and mostly formed in combustion processes of organic materials. The PAHs sources include emissions from automobiles, industrial processes, domestic heating systems, tobacco smoking and so on [1].

Human spend more than 80% time in indoors. The quality of indoor air has an important impact on human health. However, there are many sources of PAHs in indoor air, such as natural gas heating/cooking and wood or electric stoves [2]. One significant source of PAHs indoors is environmental tobacco smoke (ETS) [3]. The California Air Resources Board [4] found that the concentrations of PAHs from cigarette smoking were 1.5–4 times higher than that of other indoor combustion sources. Because of the known carcinogenic effects of many

compounds found in ETS, there is much concern over exposure to ETS. This is especially true in the indoor environment where, because of inadequate ventilation, ETS concentrations can be many times the levels found in the outdoor ambient environment [5].

ETS consists of mainstream smoke (MS), the portion that leaves the mouth end of a cigarette; sidestream smoke (SS), the portion releases in the static burning period between puffs [6]. The composition of SS was qualitatively similar to that of MS, however it was quantitatively different, SS was the major contributor quantitatively to ETS [7]. Several reports have examined the SS to MS ratio of smoke components including selected PAHs. Depending on the type of cigarette and experimental situation, the PAHs concentrations ratio of SS to MS ranged from 2 to 20 [8]. Such results may be caused by the incomplete combustion due to the limited oxygen available to the cigarettes' fire coal during the static burning period. Many researches on PAHs pollutions in MS and SS have been conducted thus far. In an actual environment, people breathe in the mixed tobacco smoke (TS) consisting of MS and SS, however little information on PAHs concentrations and source characteristic in TS is available.

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The objectives of this study are: (1) to characterize the concentrations of selected PAHs in TS of 12 commercial brand cigarettes, and discuss the pollution patterns of PAHs in TS; (2) to investigate the contribution of benzo[a]pyrene (BaP) from TS to indoor air pollution of resident homes.

2. Experimental

2.1. Experimental chamber

The research was undertaken in an experimental chamber which was constructed with low-emitting materials, it was a 20.25 m³ room (3 × 2.5 × 2.7). Before the experiment was started, background concentrations of PAHs in chamber air had been determined, and during the experiment, all items in the chamber were isolated from outside, the indoor temperature and relative humidity were in the range of 12–17 °C and 50–60%, respectively.

2.2. Cigarette smoking

Twelve commercial brand cigarettes were tested in this study. The cigarettes were all filtered brands (all brand cigarettes had 84 mm long and 7 mm in diameter, No. 3 cigarette's filter tip was 34 mm long and others' were 24 mm long). One of the commercial brand cigarettes was from USA, the others were all Chinese brands. Before the experiment was started, the cigarettes were conditioned at 15 °C and 40% relative humidity for 24 h.

The smoking machines made by ourselves were used to generate TS, fifteen cigarettes were smoked in the chamber for each brand using three smoking machines (each machine smoked five cigarettes), the cigarette smoke was generated under ISO condition (60 s puff interval, 2 s duration, and 35 mL puff volume) [9]. The MS was generated during 2 s/min duration while the SS was generated during the smolder cycle of the smoking in 58 s/min, then the TS consisting of MS and SS diluted into the chamber air. Each cigarette was smoked for 8 min, and 48 mm in length was consumed approximately. After the machine smoking, the chamber was left for 3 min before air sampling in order to have a complete mixing of air in the chamber.

2.3. Air sampling

The TS was sampled by the air sampler (DDY-1.5, Xingyu, China). The particulate PAHs were collected with 25 mm glass fibre filters (GF, Whatman, England), which were thermally treated at 500 °C for 6 h to remove organic contaminants. The vapor PAHs were adsorbed by XAD-2 (2.5 g, Supelco, USA), which was cleaned with dichloromethane and methanol until no peak of PAHs was found in HPLC. The smoking machines and air sampler were placed in the middle of chamber, about 0.5 m above the ground. The sampling was lasted for 2 h with 1.0 L/min.

To study the impact of TS on PAHs concentrations in indoor air, eight resident homes were selected, and BaP was determined

Table 1
Description of the sampling sites

No.	Sampling sites	Area of homes (m ²)	Environmental conditions
A1	Bedroom	26	Nonsmoking
A2	Bedroom	15	Smoking
B1	Living room, bedroom, kitchen	40	Nonsmoking
B2	Living room, bedroom, kitchen	40	Smoking
C1	Living room, bedroom, kitchen	65	Nonsmoking
C2	Living room, bedroom, kitchen	65	Smoking
D1	Living room, bedroom, kitchen	93	Nonsmoking
D2	Living room, bedroom, kitchen	93	Smoking

in indoor air during summer and autumn, respectively. In six of the eight, each of the measured homes had air samplers in living room, bedroom, and kitchen. The two others only had an air sampler in living room, because the rooms were very small (Table 1). The doors and windows to outside were closed, and doors of other rooms were open during air sampling. Indoor air was sampled by the air sampler (MP-15CF mini pump, Shibita, Japan). The same glass fibre filters and XAD-2 were used to collect particulate and vapor PAHs, respectively. The samplers were placed to a height of 1.5 m above the ground, and the sampling was lasted for 12 h with 1.0 L/min. The flow rates were measured before and after the sampling programs, and all deviations of the rates were less than 1%.

2.4. Analysis of PAHs samples

The determination of PAHs in the particulate and vapor phases in air was described in previous paper [1]. In general, after air sampling, the glass fibre filters were cut into pieces, placed in a 25 mL glass stoppered tube with 10 mL dichloromethane, XAD-2 were poured into a 25 mL glass stoppered tube with 20 mL mixture of dichloromethane and acetonitrile (v/v = 3/2). Then the samples were sonicated for 30 min. During the sonication, the water in the ultrasonic bath was replaced frequently to prevent overheating. Then 5 mL extracts of glass fibre filter and 10 mL extracts of XAD-2 were transferred into other glass stoppered tubes. Extracts with 30 µL dimethyl sulfoxide (DMSO) were evaporated under a gentle flow of nitrogen gas at room temperature and then was added to 1 mL methanol.

After considering their toxicity the following 17 PAHs were determined: naphthalene (NA), acenaphthene (AC), acenaphthylene (ACY), fluorene (FLUOR), phenanthrene (PHEN), anthracene (AN), fluoranthene (FLUR), pyrene (PY), benzo[a]anthracene (BaA), chrysene (CHRY), benzo[e]pyrene (BeP), 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). PAHs were determined by HPLC (Agilent, 1100 series, USA) containing a column (Vydac, C18, 250 mm × 4.6 mm), and a UV detector (Agilent, G1314A, VWD).

PAHs recovery studies were undertaken to demonstrate the availability of the analytical method. The recoveries of 17 PAHs for particulate phase and vapor phase ranged from 85% to 100% except for NA (its recoveries were 78% and 76% for glass fibre

filters and XAD-2, respectively), and the relative standard deviations of the recoveries of 17 PAHs were less than 3.7%.

3. Results and discussion

3.1. Concentrations of PAHs in tobacco smoke

The PAHs concentrations in vapor phase and particulate phase in TS are presented in Table 2. In this study, the determined PAHs covered a range from two rings which were mainly in vapor phase to six rings which were mainly in particulate phase in ambient air [10]. The sum of 17 PAHs concentrations in vapor phase and particulate phase were 3500 and 1152 ng/m³, respectively. PAHs in TS were mostly in vapor phase (accounted for 75.2% of total PAHs), but the proportion of vapor phase PAHs in TS was lower than that in ambient air [11]. Among 17 PAHs, NA was the most abundant one in vapor phase with 1824 ng/m³ in chamber, followed by FLUOR and ACY with 459.5 and 412.7 ng/m³, respectively; however, in particulate phase, the most abundant one was BP with 192.4 ng/m³, followed by PHEN and FLUOR with 139.9 and 130.1 ng/m³, respectively. BaP has been known to have strong carcinogenicity to human, in this study, the average concentration of BaP in TS was 46.87 ng/m³ (2.005 ng/m³ in vapor phase and 44.86 ng/m³ in particulate phase), it was much higher than that determined in commonly ambient air [12]. No. 3 cigarette had a long filter tip, 17 PAHs (\sum PAHs) and BaP concentration in its TS were 2270 and 23.41 ng/m³, respectively (Table 3). The concentrations were lower in NO. 3 cigarettes than all other cigarettes. The results indicated that filter tip could reduce PAHs concentrations in TS effectively. \sum PAHs and BaP concentration in TS of an American brand cigarette (NO. 8) were 2458 and 35.19 ng/m³, respectively, their concentrations were lower than that in TS of most Chinese cigarettes.

Table 2
Concentrations of 17 PAHs in tobacco smoke of 12 commercial brand cigarettes in a chamber (ng/m³)

PAHs	Vapor phase	Particulate phase	Total
Naphthalene	1824	47.09	1871
Acenaphthene	216.3	19.97	236.3
Acenaphthylene	412.7	42.01	454.7
Fluorene	459.5	130.1	589.6
Phenanthrene	150.0	139.9	289.9
Anthracene	24.71	45.15	69.86
Fluoranthene	195.7	87.38	283.1
Pyrene	89.07	90.59	179.7
Benzo[a]anthracene	4.659	40.16	44.82
Chrysene	7.871	30.89	38.76
Benzo[e]pyrene	84.52	116.6	201.1
Benzo[b]fluoranthrene	3.211	50.33	53.54
Benzo[k]fluoranthrene	12.89	28.87	41.76
Benzo[a]pyrene	2.005	44.86	46.87
Dibenzo[a,h]anthracene	0.776	11.12	11.90
Benzo[ghi]perylene	11.07	192.4	203.4
Indeno(1,2,3-cd)pyrene	1.317	34.92	36.24
\sum PAHs	3500	1152	4652

Values were acquired by averaging the data for 12 brand cigarettes.

Table 3

Smoking tar and nicotine contents (mg) in each brand cigarette, PAHs and BaP concentrations (ng/m³) in tobacco smoke of each brand cigarette

Cigarette no.	Smoking tar	Nicotine	\sum PAHs	BaP
1	15	1.3	5197	42.98
2	15	1.2	12802	43.42
3 ^a	15	1.3	2270	23.41
4	15	1.2	2961	49.27
5	8	0.7	2465	48.46
6	15	1.2	2471	43.66
7	17	1.3	3251	66.48
8 ^b	12	1.0	2458	35.19
9	15	1.0	5353	69.83
10	15	1.2	5134	41.90
11	15	1.2	5512	44.69
12	10	1.0	5955	53.07

^a No. 3 cigarette had a long filter tip.

^b No. 8 was an American cigarette, others were Chinese cigarettes.

3.2. Patterns and fingerprint of PAHs in tobacco smoke

The proportion of PAHs in particulate phase proportion increased with increasing molecular weight (Fig. 1). It appears that the molecular weight of PY is the dividing line for the PAHs with a larger or smaller distribution in the vapor or particulate phase. PY has 49.6% in vapor phase, and 50.4% in particulate phase. For PAHs with higher molecular weights, the percentage in particulate phase becomes significantly higher. An exception of this is AN, which has 35.4% in vapor and 64.6% in particulate. However, compared to ambient air [1], the particulate phase proportion of three and four rings PAHs such as PHEN, AN, PY was higher in TS than that in ambient air, and the vapor phase proportion of five and six rings PAHs such as BeP, BkF was also higher in TS than that in ambient air. So, the results indicated that the distribution of PAHs in vapor phase and particulate phase was closer in TS than that in ambient air.

The distribution of PAHs with different rings in TS is presented in Fig. 2. With the rings increasing, the proportions of different rings PAHs to \sum PAHs decreased. It was accounted for

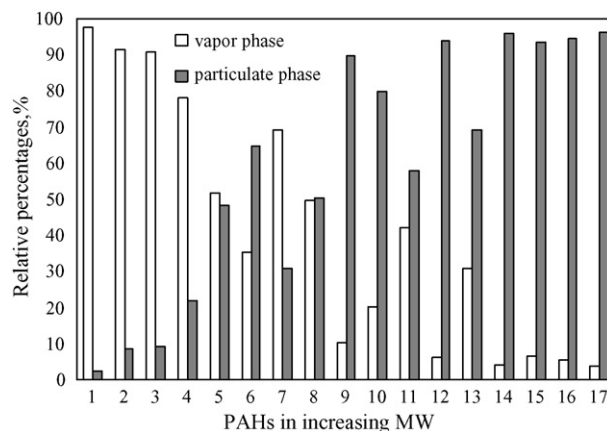


Fig. 1. Distribution of PAHs in vapor phase and particulate phase in tobacco smoke (1–17 on abscissa stand for NAPH, AC, ACY, FLUOR, PHEN, AN, FLUR, PY, BaA, CHRY, BeP, BbF, BkF, BaP, DA, BP, IN, respectively. Figure was acquired by averaging the data for 12 brand cigarettes).

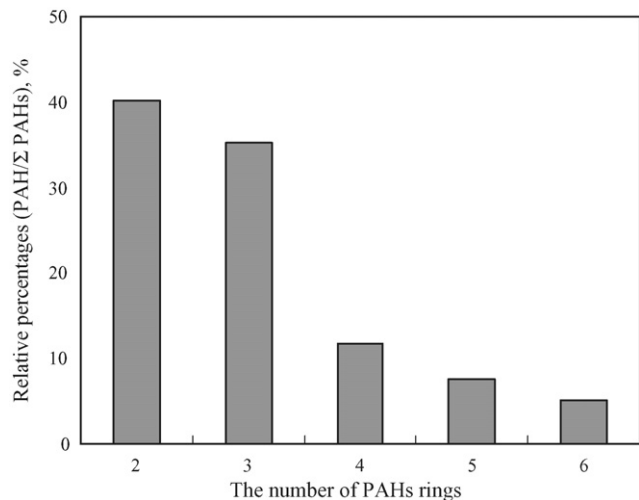


Fig. 2. Distribution of PAHs with different rings in tobacco smoke (figure was acquired by averaging the data for 12 brand cigarettes).

40.2%, 35.3%, 11.7%, 7.6%, 5.2% for two to six rings PAHs, respectively. This distribution was much different as found in cooking oil-fume, automobile exhaust and exhaust gas of coke plant [13–15].

The smoking tar and nicotine are also harmful substances in cigarette to human health. They are regarded as the important indexes in cigarette manufacture processes. The smoking tar and nicotine in 12 brand cigarettes are presented in Table 3. The correlation analysis was made for PAHs concentrations to smoking tar contents and nicotine contents, BaP concentrations to smoking tar contents and nicotine contents, there were no obvious correlations for PAHs concentrations to smoking tar contents ($r=0.15$, $p>0.05$), PAHs concentrations to nicotine contents ($r=0.14$, $p>0.05$), BaP concentrations to smoking tar contents ($r=-0.11$, $p>0.05$), and BaP concentrations to nicotine contents ($r=0.30$, $p>0.05$). In recent years, the chemical mass balance (CMB) models have been developed to understand the relationship between PAHs sources and their concentrations in air. CMB models use the chemical and physical characteristics of sources and receptors to both identify the presence of source and quantify the source contribution to the receptor [16]. TS is not only a significant source of PAHs in indoor air, but also a source in ambient air of some public place, so a source fingerprint of PAHs in TS is a crucial requirement of CMB models to identify and quantify TS contribution to PAHs in air. The source fingerprint of PAHs in TS is shown in Fig. 3. NA, regarded as the marker of TS source, had the largest contribution to \sum PAHs (40.2%), followed by FLUOR (12.7%) and ACY (9.8%), and other PAHs contributions to \sum PAHs were all under 7%.

3.3. PAHs yields of cigarette in tobacco smoke

The PAHs yields are also presented in Table 4. The average yields of vapor phase PAHs and particulate phase PAHs were 4725 and 1555 ng/cig, respectively. BaP yield was 2.707 ng/cig in vapor phase and 60.56 ng/cig in particulate phase. Few literatures on simultaneously determining vapor phase PAHs and particulate phase PAHs were available. Ding [6] determined 14

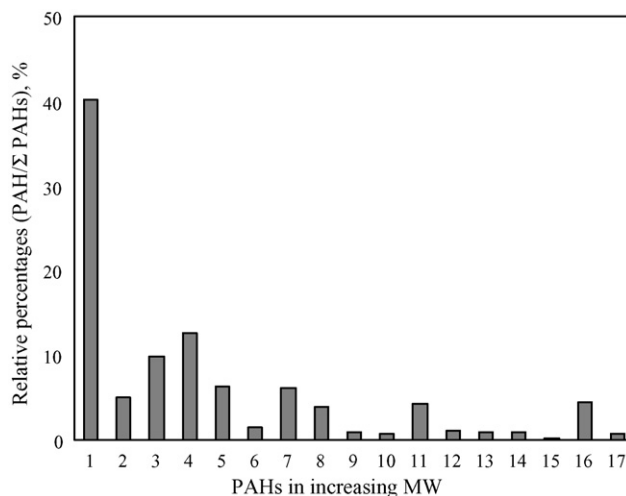


Fig. 3. PAHs fingerprint for tobacco smoke (1–17 on abscissa stand for NAPH, AC, ACY, FLUOR, PHEN, AN, FLUR, PY, BaA, CHRY, BeP, BbF, BkF, BaP, DA, BP, IN, respectively). Figure was acquired by averaging the data for 12 brand cigarettes).

PAHs in MS with the range from 87 to 1598 ng/cig in particulate phase. Baek and Jenkins [7] determined 16 PAHs in SS with the average yields of 2290 ng/cig in particulate phase for commercial cigarette, in which BaP yield was 215 ng/cig in particulate phase. The difference of the results between this study and literatures may be associated with two possible reasons. First, the cigarette's materials and manufacturing processes could be different in different countries, this would be surely result in the difference of PAHs yields. Many sorts of ingredients were added to raw tobacco to reduce the tar levels, one kind was plant ingredient, it would make greater evaporation of more volatile components and tended to increase the relative concentrations of semi-volatile components such as BaP when smoking. Second, different smoke generating protocols in different

Table 4
PAHs yields of cigarette in tobacco smoke (ng/cig)

PAHs	Vapor phase yields	Particulate phase yields	Total yields
Naphthalene	2462	63.57	2526
Acenaphthene	292.1	26.96	319.1
Acenaphthylene	557.2	56.71	613.9
Fluorene	620.4	175.6	796.0
Phenanthrene	202.5	188.8	391.3
Anthracene	33.36	60.95	94.30
Fluoranthene	264.2	118.0	382.2
Pyrene	120.2	122.3	242.5
Benzo[a]anthracene	6.290	54.22	60.51
Chrysene	10.63	41.70	52.32
Benzo[e]pyrene	114.1	157.4	271.5
Benzo[b]fluoranthrene	4.335	67.94	72.28
Benzo[k]fluoranthrene	17.40	38.98	56.38
Benzo[a]pyrene	2.707	60.56	63.27
Dibenzo[a,h]anthracene	1.048	15.02	16.07
Benzo[ghi]perylene	14.95	259.7	274.6
Indeno(1,2,3-cd)pyrene	1.778	47.14	48.92
\sum PAHs	4725	1555	6281

Values were acquired by averaging the data for 12 brand cigarettes.

Table 5
Concentrations of BaP in eight homes (ng/m³)

No.	Environmental condition	Concentrations of BaP (summer)	Concentrations of BaP (autumn)
A1	Nonsmoking	3	4
A2	Smoking	11	12
B1	Nonsmoking	11	17
B2	Smoking	12	20
C1	Nonsmoking	9	9
C2	Smoking	17	17
D1	Nonsmoking	2	4
D2	Smoking	15	21
Mean ± S.D.		10.0 ± 5.3	13.0 ± 6.8

countries, difference in experimental chamber configuration (e.g., different temperature and humidity), difference in TS sampling (e.g., different smoking duration for each cigarette), could make the difference of the results between this study and literatures.

3.4. Contribution of BaP from tobacco smoke to indoor air pollution of resident homes

Because of BaP's strong and direct carcinogenicity, several studies focused on its concentrations in ETS [17,18]. It was also reported that BaP concentration of indoor air in a heavy smoker's homes was about 10 times higher than that in a nonsmoker's home [19].

The BaP concentrations of indoor air in nonsmoking homes were from 2 to 11 ng/m³ in summer; the corresponding levels in smoking homes were from 11 to 17 ng/m³. In autumn, its concentrations in nonsmoking and smoking homes were from 4 to 17 and 12 to 21 ng/m³. The average BaP concentrations in resident homes were 10 ng/m³ in summer and 13 ng/m³ in autumn, respectively (Table 5). The BaP concentration of indoor air in Hangzhou was much higher than that in Shizuoka, where the mean was 1.6 ng/m³ reported in our previous study [20].

Statistical analysis (*t*-test) was performed to analyze difference between BaP concentrations in smoking and nonsmoking resident homes. The result indicated that BaP concentration of indoor air in smoking homes was significantly higher than that in nonsmoking homes ($p < 0.01$). In this study, the selected eight homes were four contrasting pairs which were in the same floor of the same building, had similar construction structures and living habits except for smoking. So, it was concluded that ETS made a great contribution to BaP in indoor air. The D1 and D2 homes were in suburb of Hangzhou, and there were fewer pollution sources in outdoor air, so BaP level in D1 home can be seen as the background level of BaP for D2 home, the contribution of smoking to BaP of the D2 home were 87% and 81% in summer and autumn, respectively.

4. Conclusions

Environmental tobacco smoke is a significant source of PAHs in indoor air. In order to find out the pollution patterns of

PAHs in tobacco smoke, concentrations of 17 PAHs in tobacco smoke of 12 commercial brand cigarettes were determined by simulated experiment in a chamber. The source fingerprint of PAHs in tobacco smoke was established by the experiment, with the source fingerprint, NA could be regarded as the marker of tobacco smoke source because of its largest contribution to \sum PAHs (40.2%), followed by FLUOR (12.7%) and ACY (9.8%). Further study indicated that more than 80% of BaP in indoor air of resident homes in Hangzhou was from tobacco smoke.

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