

Characteristics of polycyclic aromatic hydrocarbons in PM_{2.5} emitted from different cooking activities in China

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Abstract Nineteen polycyclic aromatic hydrocarbons (PAHs) in PM_{2.5} emitted from five different cooking activities were characterized, and their influencing factors were determined. The total quantified particle-bounded PAH concentrations (Σ PAHs) in the air from the cooking activities were 4.2–36.5-fold higher than those in corresponding backgrounds. The highest Σ PAHs were seen in cafeteria frying (783 ± 499 ng/m³), followed by meat roasting (420 ± 191 ng/m³), fish roasting (210 ± 105 ng/m³), snack-street boiling (202 ± 230 ng/m³), and cafeteria boiling (150 ± 65 ng/m³). The main influencing factors on the PAH emissions were cooking methods, fat contents in raw materials, and oil consumptions. Four- to six-ringed PAHs had the highest contributions to the Σ PAHs (avg. 87.5%). Diagnostic

ratios of individual PAH were similar between the two charbroiling and other three conventional Chinese cooking methods, respectively, demonstrating the dominance of cooking methods in the PAH emissions. Remarkably high benzo(b)fluoranthene/benzo(k)fluoranthene (BbF/BkF) ratio (8.31) was seen in the snack-street boiling, attributed to the coal combustion as cooking fuel. Both fluoranthene/(fluoranthene + pyrene) [FLT/(FLT + PYR)] and benzo(a)anthracene/(benzo(a)anthracene + chrysene) [BaA/(BaA + CHR)] ratios were higher for the oil-based cooking than those from the water-based ones. In addition, two ratios of indeno(1,2,3-cd)pyrene/(indeno(1,2,3-cd)pyrene + benzo(g,h,i)perylene) [IPY/(IPY + BPE)] and benzo(a)pyrene/(benzo(a)pyrene + benzo(g,h,i)perylene) [BaP/(BaP + BPE)] were higher for two charbroiling than the three conventional Chinese cooking methods. The characterization work in this study is particularly important since cooking is a potential contributor of atmospheric PAHs in urban China. Carcinogenic potencies of PAHs were assessed by comparison with the air quality guideline and health risk estimation. The BaP and BaP equivalent were higher for the oil-based than the water-based cooking activities.

Yun-Chun Li and Jia-Qian Qiu contributed equally to this work.

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Introduction

Fine particulate matter (particulate matter of aerodynamic diameters of less than 2.5 μ m (PM_{2.5})) has adverse effect to human health with an increasing risk of morbidity and mortality from cardiovascular and respiratory diseases. The particles also cause visibility deterioration in ambient environments (Harrison and Yin 2000; Pui et al. 2014; Yadav et al. 2003). The major sources of PM_{2.5} in China include coal

combustion, vehicle emission, biomass combustion, industry and road dust (Hu et al. 2010; Liu et al. 2015; Pui et al. 2014). Several studies illustrated that cooking is an important source of ambient $PM_{2.5}$, particularly in populated urban areas (Allan 2009; Mohr et al. 2012; Schauer et al. 2007). Meat cooking discharged about 11.6 tons PM daily, and meat charbroiling and frying accounted for approximately 7% of the total $PM_{2.5}$ masses in the atmosphere of Los Angeles, USA (Hildemann et al. 1991; Schauer et al. 2007). Cooking emission is also a significant contributor to atmospheric organic carbon (OC). The contribution of meat cooking to ambient OC in fine PM ranged from 15 to 23% (Brinkman et al. 2009; Hildemann et al. 1991; Rogge et al. 1991; Schauer et al. 1999, 2007; Watson et al. 1998). Besides, the pollutants discharged from the cooking activities have threat to human health due to their high potentials of lung cancer. The cooking oil fumes contain harmful substances such as polycyclic aromatic hydrocarbons (PAHs) (e.g., benzo[a]pyrene (BaP), dibenzo[a,h]anthracene (DBA), benzo[b]fluoranthene (BbF), and benzo[a]anthracene (BaA)). More attentions on the cooking emissions have been thus drawn with their carcinogenic, teratogenic, and mutagenic toxicity (Chiang et al. 1997; Ko et al. 1997; Liu et al. 2009; Qu et al. 1992; See and Balasubramanian 2006; Wu et al. 1998, 1999; Zhong et al. 1999).

Generated by incomplete combustion and/or pyrosynthesis of organic materials, atmospheric PAHs and their derivatives are a large group of chemicals with two to seven fused aromatic rings. Light PAHs (with less than four aromatic rings) mainly exist in the gas phase because of high vapor pressures, while heavier PAHs present in the particle phase or scatter in the atmospheric soot. Higher carcinogenicity of a PAH was generally reported with an increasing of its molecular weight (Akyüz and Cabuk 2010; Ravindra and Grieken 2008).

Each cooking activity has distinguished emission characteristics. The PAH emissions could be affected by many factors, such as cooking methods, oil consumptions, fat contents in foods, and fuel types and temperatures (Chen and Chen 2015; Rogge et al. 1991). Rogge et al. (1991) reported that preparation of charbroiled-meat hamburger released more PAHs (avg. 2.56 mg/kg) than fried-meat hamburger (avg. 0.35 mg/kg). In addition, the amounts of PAHs released from deep frying (oil-based cooking) were 2.6 and 3.5 times of those from boiling and steaming (water-based cooking), respectively (See and Balasubramanian 2008). Yao et al. (2015) also found that deep frying generated more PAHs (including BaP) than typical frying. Similarly, frying emitted more PAHs than boiling for low-fat foods (Zhu and Wang 2003). Saito et al. (2014) and Tanaka et al. (2012) concluded that the fat contents of foods and ingredients significantly varied the PAH formations during thermal cooking processes. This could be further supported by the case that preparation of chicken nuggets and hairtails produced more PAHs than potato and eggs because of their higher fat contents (Yao et al. 2015). Cooking

fuels contribute on the PAH production. For instance, electricity and natural gas combustion release less PM and PAHs than those from coal combustion and biomass burning (Amouei et al. 2017; Park et al. 2011; Ravindra and Grieken 2008; Shen et al. 2013).

Among those typical cooking methods, the Chinese styles are characterized by distinctive cooking materials, cooking techniques and flavors, and seasoning. The amounts of PAH emissions were much less from Western fast food cooking than the Chinese cooking. Chrysene (CHR) and pyrene (PYR) were the two major PAH components in Western-style cooking emissions, while PYR, fluoranthene (FLT), and benzo(g,h,i)perylene (BPE) were the dominant species from the Chinese cooking (Zhao et al. 2007). CHR was the most important PAH in meat charbroiling (Rogge et al. 1991), and PYR had the highest contribution in Chinese cooking (He et al. 2004). Even though many researches have been conducted on the PM emissions from Chinese cooking, significant variations might exist due to variable operation conditions. The main objectives of this study are to investigate the PAH emission characteristics in $PM_{2.5}$ generated from five common local cooking activities and determine their influence factors. The results could offer scientific data on the cooking emissions, which are informative for establishing effective strategies for air quality control and health protection in China.

Experimental section

Sample collection

$PM_{2.5}$ samples generated from five cooking activities were collected in Yucheng City, Ya'an, China from August to mid-September 2013. Detailed sampling information was reported in our previous study (Li et al. 2015) and shown in Table 1. Briefly, the sampling events were conducted on a commercial street (Xinkang Street) and at Sichuan Agricultural University (SAU). Two background sites were selected for comparison. A street background site was assigned on Xinkang Street, where it is close to a barbecue restaurant and the Wanzhou grill fish restaurant. A campus background site was set up on the roof of an 11-story teaching building, which is about 300 m away from the cafeteria and snack street on the campus. $PM_{2.5}$ was collected onto quartz fiber filters (90 mm diameter, Staplex, USA), pre-baked at 500 °C for 8 h, with a KC-120H medium-volume sampler (Qingdao Laoshan Electronic Instrument Factory Co., Ltd., China) operated at a flow rate of 100 L/min. The filter holders and open ends were cleaned with deionized water after each round of sampling. The sample-loaded filters were properly stored in a refrigerator at -20 °C. All samples were collected during business hours for 2–4 h, subjected to the flow of

Table 1 Sampling information for the five cooking activities

Category	Outdoor charbroiling		Conventional Chinese cooking		
	Meat roasting	Fish roasting	Snack-street boiling	Cafeteria frying	Cafeteria boiling
Scale	Small-scale (0 ~ 50 persons)	Small-scale (0 ~ 50 persons)	Middle-scale (100 ~ 300 persons)	Large-scale (> 500 persons)	Large-scale (> 500 persons)
Sampling place	1.5 m apart from the grills in a barbecue restaurant in Xinkang Street	1.5 m apart from the grills in Wanzhou grill fish restaurant in Xinkang Street	Dining area in a noodle restaurant in snack street of SAU	1.0 m apart from the chimney outlet from a frying cafeteria in SAU	1.0 m apart from the chimney outlet from a boiling cafeteria in SAU
Sampling period	21:30–01:30	17:00–21:00	11:00–14:00, 16:00–20:00	10:30–13:00, 16:00–18:30	10:30–13:00, 16:00–18:30
Fuel type	Charcoal	Charcoal	Natural gas, coal	Natural gas	Natural gas
Cooking style	Roasting	Roasting	Boiling, frying (around the sampling place)	Frying (pan frying, stir frying, deep frying)	Boiling
Ingredient and property	Mutton, chicken, pork, beef, seafood, vegetables, 5-spice powder; more oil based	Fish, much salt; less oil based	flour (noodles, wontons), water based	Meat, vegetables, 5-spice powder; more oil based	Vegetables, flour (noodles, wontons), water based

diners. The background samples were obtained for 4 h when no cooking activity was conducted. Four field blanks were simultaneously collected as well. A total of valid 44 filter samples were collected in this study.

PM_{2.5} mass, OC, and EC analyses

PM_{2.5} mass was obtained from the weight difference of a filter sample prior to and after sampling. Before weighting, the filters were equilibrated in a controlled chamber (temperature, 30 °C; relative humidity, 30–60%) for 24 h.

A filter punch of 0.526 cm² in size from each filter was obtained for elemental carbon (EC) and OC analyses using a DRI Model 2001 Thermal/Optical Carbon Analyzer (Atmoslytic Inc., Calabasas, CA, USA). The quartz filter was analyzed for four OC and three EC fractions following the IMPROVE_A thermal/optical reflectance protocol (Chow et al. 1993). The analyzer was calibrated with known quantities of CH₄ daily. Detailed information on carbon analysis and required quality assurance and quality control (QA/QC) were documented in Cao et al. (2003).

PAH analysis

Nineteen target PAHs were analyzed by thermal desorption-gas chromatography-mass spectrometry (TD-GC-MS) using an Agilent 7890A/5975C system (Ho and Yu 2004; Ho et al. 2008, 2011) (Table 2). Punches of quartz-fiber filter samples (0.526 or 1.052 cm²) were spiked with internal standards (*n*-

C₂₄D₅₀ and Phe-d₁₀). Each filter piece was placed into the Pyrex glass TD tube (78 mm long, 4 mm i.d., and 6 mm o.d.). Pre-baked glass wool was used as a plug to hold the filter parts in position. The sample loaded tube was exchanged into the GC injector port when its temperature was lower to 50 °C for analysis, and then manually raised to 275 °C for desorption in a splitless mode after the injector cap was closed. An HP-5ms capillary column (30 m × 0.25 mm × 0.25 μm, J&W Scientific, Folsom, CA, USA) was used to separate the analytes. The temperature program for the GC oven was as follows: initially held at 30 °C for 2 min, ramped to 120 °C at 10 °C/min, then ramped to 310 °C at 8 °C/min, and finally held at 310 °C for 20 min. A constant carrier gas (helium) flow was maintained at 1.0 mL/min. The MS was operated in scan mode from 50 to 650 amu. The ion source temperature was 230 °C, and a voltage of 70 eV was applied for the electron impact ionization.

Data analysis

Several PAH are known as human carcinogens. BaP is of primary concern because of the highest carcinogenicity. The concentration of BaP could be used to assess toxicity. In addition to BaP, high molecular mass PAHs, such as BaA, BbF, BkF, IPY, and DBA have high carcinogenicity. Therefore, BaP equivalent carcinogenic power (BaPE), based on carcinogenic potency relative to BaP, was also used to estimate the exposure risks for the six PAHs with an equation shown below (Liu et al. 2009; Yassaa et al. 2001):

Table 2 Mass concentrations of PM_{2.5}, OC, EC, and 19 PAHs

Species	Meat roasting <i>N</i> = 7	Fish roasting <i>N</i> = 7	Street background <i>N</i> = 6	Snack-street boiling <i>N</i> = 6	Cafeteria frying <i>N</i> = 3	Cafeteria boiling <i>N</i> = 9	Campus background <i>N</i> = 6
Mass (µg/m ³)							
PM _{2.5}	1107 ± 340 ^a	493 ± 176	152 ± 23	329 ± 68	992 ± 192	257 ± 29	104 ± 17
OC	655 ± 250	185 ± 84	14.7 ± 2.4	42.8 ± 6.8	523 ± 88	66.7 ± 22.7	10.7 ± 5.8
EC	12.0 ± 2.4	11.3 ± 4.8	2.18 ± 0.44	2.61 ± 0.90	9.93 ± 2.35	7.17 ± 1.44	1.90 ± 1.57
PAHs (ng/m ³)							
Acenaphthene (ACE) ^b	ND ^c	ND	ND	ND	77.2 ± 40.0	ND–244.5 ^d	ND
Fluorene (FLU)	ND	ND–2.1	ND	ND	12.3 ± 5.5	ND–1.0	ND
Phenanthrene (PHE)	ND–18.2	ND	ND	ND	38.9 ± 34.2	ND	ND
Anthracene (ANT)	13.5 ± 8.7	0.9 ± 0.4	ND–0.5	ND	2.5 ± 2.0	1.1 ± 2.7	ND–1.5
Fluoranthene (FLT)	53.6 ± 33.6	15.3 ± 10.6	ND–1.3	ND	74.7 ± 46.2	4.4 ± 3.6	2.1 ± 2.6
Pyrene (PYR)	51.9 ± 32.1	14.5 ± 11.5	ND–1.1	ND–0.5	159 ± 126	7.5 ± 4.9	2.2 ± 3.0
Benzo(a)anthracene (BaA)	31.3 ± 13.1	17.1 ± 9.3	ND–1.0	6.8 ± 9.4	13.4 ± 5.7	4.6 ± 5.2	1.0 ± 1.3
Chrysene (CHR)	37.7 ± 13.9	26.7 ± 17.6	ND–0.8	15.0 ± 20.3	15.2 ± 4.7	4.0 ± 1.1	2.4 ± 1.9
Benzo(b)fluoranthene (BbF)	17.6 ± 9.9	18.5 ± 8.1	2.7 ± 1.3	28.3 ± 25.2	18.1 ± 6.9	9.9 ± 2.4	5.6 ± 3.2
Benzo(k)fluoranthene (BkF)	36.9 ± 14.1	22.1 ± 13.7	2.1 ± 0.6	6.4 ± 8.8	13.2 ± 5.6	10.1 ± 7.2	4.3 ± 2.6
Benzo(a)fluoranthene (BaF)	35.3 ± 14.1	5.7 ± 1.2	1.3 ± 0.1	1.6 ± 1.7	3.3 ± 0.4	5.9 ± 7.7	2.3 ± 0.7
Benzo(e)pyrene (BeP)	30.6 ± 10.7	22.4 ± 11.2	2.1 ± 1.2	69.0 ± 94.3	44.9 ± 34.7	18.4 ± 7.7	5.3 ± 4.4
Benzo(a)pyrene (BaP)	25.8 ± 10.6	12.2 ± 4.9	1.2 ± 0.6	5.8 ± 7.8	24.7 ± 22.9	7.3 ± 4.6	2.9 ± 2.8
Perylene (PYL)	23.2 ± 9.1	5.1 ± 0.8	0.5 ± 0.1	1.6 ± 1.7	5.5 ± 5.0	4.5 ± 5.1	0.9 ± 0.5
Dibenzo(a,h)anthracene (DBA)	8.9 ± 3.3	2.4 ± 0.6	ND	10.3 ± 15.3	0.1 ± 0.1	1.7 ± 2.4	ND–0.5
Indeno(1,2,3-cd)pyrene (IPY)	24.1 ± 9.2	11.9 ± 5.5	0.6 ± 0.4	4.4 ± 5.6	11.2 ± 7.3	7.1 ± 5.2	1.7 ± 1.9
Benzo(g,h,i)perylene (BPE)	11.5 ± 4.1	21.9 ± 13.8	1.0 ± 1.1	28.4 ± 34.2	172 ± 171	40.1 ± 33.6	3.9 ± 4.7
Dibenzo(a,e)pyrene (DBP)	9.4 ± 3.5	3.5 ± 1.4	ND	15.0 ± 21.5	4.1 ± 4.8	1.7 ± 2.0	ND–0.3
Coronene (COR)	9.0 ± 3.4	9.7 ± 5.6	ND–0.5	10.0 ± 10.8	92.5 ± 90.7	21.9 ± 16.8	1.0 ± 1.4
ΣPAHs	420 ± 191	210 ± 105	11.5 ± 5.3	202 ± 230	783 ± 499	150 ± 65	35.6 ± 29.5

^a Means average concentration ± one standard deviation

^b The abbreviation of individual PAH compound is shown in the bracket

^c ND represents below detection limit and/or below field blank

^d Only range was reported for > 50% of data below detection limit and/or below field blank. Their average and standard deviation are not calculated

$$BaPE = 0.06 BaA + 0.07 B[b + k]F + BaP + 0.6 DBA + 0.08 IPY \quad (1)$$

where both PAH concentrations and calculated units are in nanograms per cubic meter. Besides, the carcinogenic potency of total quantified PAHs can be calculated by toxic equivalent factor (TEF). TEF represents the relative carcinogenic potency of given PAH compound, which use BaP as a reference to adjust its original concentration (Barbosa et al. 2006; Li et al. 2003; Liu et al. 2009; Yao et al. 2015). In this study, TEF list provided by Nisbet and LaGoy (1992) (Table S1, Table S# denotes materials provided as supporting information) was used which reflects well the actual knowledge of the toxic potency of each individual PAH compound. The BaP toxic equivalent quantity (TEQ) of all considered PAHs could be calculated:

$$TEQ = \sum C_{x,TEQ} = \sum C_x \times F_x \quad (2)$$

where $C_{x,TEQ}$ is the TEQ concentration (ng/m³) of component x , which is normalized using the toxicity of BaP, C_x is the original concentration (ng/m³) of component x , and F_x is the TEF of component x . Hereby, the carcinogenic potency of total PAHs can be assessed by TEQ.

Results and discussion

PM_{2.5} and its components from different cooking activities

Table 2 shows the mass concentrations of PM_{2.5}, OC, EC, and target PAHs in both cooking activities and background

samples. The $PM_{2.5}$ concentrations of the cooking samples were 2.5–9.6 times higher than those of corresponding backgrounds. The five cooking activities can be divided into two groups, namely, outdoor charbroiling (meat roasting and fish roasting) and conventional Chinese cooking (cafeteria frying, snack-street boiling, and cafeteria boiling), based on the fuel types and cooking methods (Li et al. 2015). Generally, the $PM_{2.5}$ concentration from the outdoor charbroiling (avg. $800 \pm 425 \mu\text{g}/\text{m}^3$) was much higher than that of conventional Chinese cooking (avg. $403 \pm 288 \mu\text{g}/\text{m}^3$). The highest OC level was found in meat roasting ($655 \pm 250 \mu\text{g}/\text{m}^3$), followed by cafeteria frying ($523 \pm 88 \mu\text{g}/\text{m}^3$), fish roasting ($185 \pm 84 \mu\text{g}/\text{m}^3$), cafeteria boiling ($66.7 \pm 22.7 \mu\text{g}/\text{m}^3$), and snack-street boiling ($42.8 \pm 6.8 \mu\text{g}/\text{m}^3$). The trend was consistent with the previous findings that the factors such as cooking method, oil consumption, fat content, and fuel type, could greatly impact on the emissions (Amouei et al. 2017; Li et al. 2015; See and Balasubramanian 2006; Wang et al. 2017).

Different from the trend of OC, the highest total quantified particle-bounded PAH concentrations (ΣPAHs) was obtained in cafeteria frying ($783 \pm 499 \text{ ng}/\text{m}^3$), followed by meat roasting ($420 \pm 191 \text{ ng}/\text{m}^3$), fish roasting ($210 \pm 105 \text{ ng}/\text{m}^3$), snack-street boiling ($202 \pm 230 \text{ ng}/\text{m}^3$), and cafeteria boiling ($150 \pm 65 \text{ ng}/\text{m}^3$) (Table 2). On a whole, the oil-based cooking methods and higher fat contents of foods and ingredients produced more PAHs. Although meat roasting had the largest emissions of $PM_{2.5}$ and OC, its ΣPAHs were lower than that produced from cafeteria frying. This could be ascribed to the cooking method and water content in foods. Charbroiling directly heated the meats over burnt charcoal, while the conventional Chinese cooking prepared the foods in boilers or on pans over stoves. During charbroiling, the fats and oils fell onto the hot charcoals and were pyrolyzed. This can lead to formation of PAHs, which were then volatilized and partly redeposited on the meat surfaces (Rogge et al. 1991). Furthermore, it was reported that charcoal-grilled meats contained the highest PAHs (Farhadian et al. 2010) which might absorb the discharged particles to some extent. In addition, the existing water in foods and ingredients could generally facilitate to produce more PAHs during cooking processes (Buonanno et al. 2011). Cafeteria frying activities not only consumed large amounts of oils, but also involved much water than roasting or charbroiling of meats. The ΣPAHs from other three cooking activities were much lower, ranged from 150 ± 65 to $210 \pm 105 \text{ ng}/\text{m}^3$. In addition, the ΣPAHs from five cooking activities were 4.2–36.5-fold higher than those in the corresponding backgrounds, demonstrating that the cooking emission had a great contribution to PAHs.

Discriminating the two roasting cooking methods, the ΣPAHs from the meat roasting ($420 \pm 191 \text{ ng}/\text{m}^3$) was approximately 2-fold of the fish roasting ($210 \pm 105 \text{ ng}/\text{m}^3$). This evidenced that the fat contents of foods and ingredients

was a critical factor. Rogge et al. (1991) observed that the meats have abundant unsaturated and saturated fatty acids, which could release organic compounds thru oxidation, decarboxylation, fragmentation, recombination, rearrangement, condensation, and cyclization reactions during thermal cooking processes. Due to higher fat content than fish, the meats generate more organic components including PAHs in charbroiling (McDonald et al. 2003). Our results are consistent with those of previous studies, concluding that the fat contents can induce a significant impact on PAH formation (Saito et al. 2014; Tanaka et al. 2012; Yao et al. 2015). Oil consumption is the next dominating factor. Due to larger amounts of oil and seasoning consumed than the fish roasting, the meat roasting emitted more PAHs. The effect of food size could not be ignored as well. The charbroiled meat was cut into small pieces, while the roasted fish was typically integrated. The small pieces promoted the surface areas where more oils could be coated. This advanced more fats exposed and temperature raised in the cooking (Li et al. 2015).

The PAHs emitted from the cafeteria frying was ~ 5.2 and 3.9 times higher than those of cafeteria and snack-street boiling, respectively. This is consistent with the results reported by See and Balasubramanian (2008), which frying (oil-based) generated more PAHs than boiling and steaming (water-based). In another study, See and Balasubramanian (2006) also found that deep frying at the Malay stall generated the highest concentration of PAHs, while the lowest was seen for boiling at the Indian stall. Compared with the water-based cooking, oil-based generally releases more PAHs due to direct evaporation, oxidation, pyrolysis, and/or degradation of organic compounds from oils at a higher temperature (Abdullahi et al. 2013; Moret and Conte 2000). When heating temperatures were above 200–300 °C, the lipids could be more liable to degrade and potentially form more aromatics (Wasserman 1972). Although the cafeteria and snack-street boiling applied the same cooking method, the latter had higher ΣPAHs . The cooking fuel of coal, bad ventilation, and other frying and boiling activities along the snack street are the possible influence factors.

Characterization of PAHs in different cooking activities

Figure 1 shows the mass compositions of individual PAH in five different cooking activities and two backgrounds. CHR, FLT, BkF, PYR, BeP, BaA, BPE, BbF, and BaP are the main PAHs released from two charbroiling activities, having an average contribution of 10.8, 10.0, 9.7, 9.6, 9.0, 7.8, 6.6, 6.5, and 6.0% of ΣPAHs , respectively. These nine compounds totally accounted for 70.6 and 81.3% of ΣPAHs for the meat roasting and fish roasting, respectively. It is noticeable that CHR had the largest composition, consistent with the results reported by Rogge et al. (1991) and Schauer et al. (1999). However, different compositions were seen for conventional

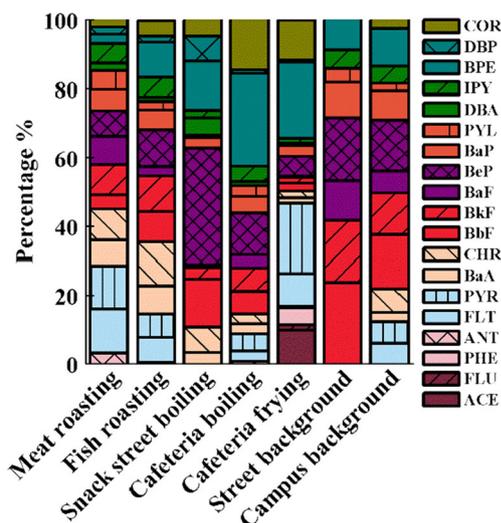


Fig. 1 Individual PAH profiles for the five cooking activities and two backgrounds

boiling in which BeP, BPE, BbF, and COR are the four dominated PAHs, averagely accounting for 23.2, 20.4, 10.3, and 9.8% of Σ PAHs, respectively. These components represented 67.0 and 60.1% of Σ PAHs for the snack-street boiling and cafeteria boiling, respectively. It is outstanding that BeP was extremely abundant in the snack-street boiling, attributed to coal used as a cooking fuel. BeP is a source marker of coal combustion (Larsen and Baker 2003). The five top PAHs from cafeteria frying were BPE, PYR, COR, ACE, and FLT, accounting for 22, 20.3, 11.8, 9.9, and 9.5% of Σ PAHs, respectively. It was found that ACE was merely detected from cafeteria frying ($77.2 \pm 40.0 \text{ ng/m}^3$), that could be explained by its rapid formation during frying under higher temperature and then condensation into the particle through the long chimney before the oil fumes were collected. In addition, Zhao et al. (2007) and He et al. (2004) also reported that PYR was the most component emitted from Chinese cooking. Both observations reveal that the constituents of PAHs could be greatly varied by different cooking activities.

On average, the six PAHs were detected in the two backgrounds, including BbF(19.7%), BeP (16.6%), BkF (15.1%), BPE (9.9%), BaP (9.1%), and BaF (9.0%). The sum of these compounds in totally accounted for 90.6 and 68.3% of Σ PAHs for the street and campus backgrounds, respectively. Among them, BbF had the largest proportion, possibly impacted by diesel emission from surrounding traffics to some degree (Park et al. 2011). Moreover, ACE, FLU, and PHE were undetectable in the backgrounds because of their low partitioning in particulate phase (Akyüz and Cabuk 2010).

Distributions of PAHs with aromatic ring numbers

Considering that the toxicity of a PAH ascends with an increase of molecular weight, the PAHs were thus classified

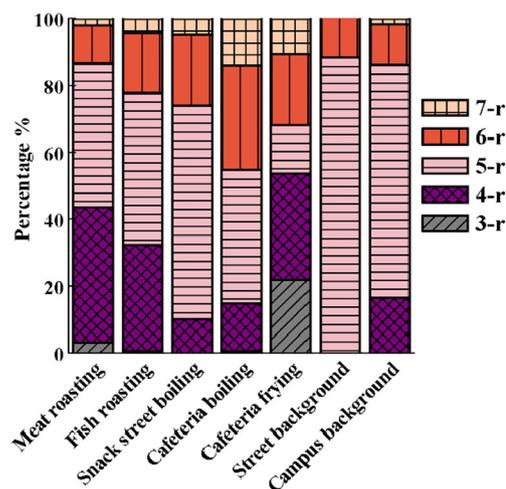


Fig. 2 Mass compositions of PAHs in terms of number of aromatic rings

with the numbers of aromatic rings (Li et al. 2003). Figure 2 illustrates the distribution of PAHs with the ring numbers from five cooking activities and two backgrounds. The total contributions of three-ringed PAHs in cafeteria frying ($21.9 \pm 13.4\%$ of Σ PAHs) were higher than those of other cooking activities ($< 3.1\%$ of Σ PAHs). In addition, the four-ringed PAHs is the major proportion for both meat roasting, fish roasting and cafeteria frying, which were accounted for 31.8 ~ 40.3% of Σ PAHs, much higher than the two water-based cooking activities of snack-street and cafeteria boiling of 10.2 ~ 14.2%. Moreover, the five-ringed PAHs were the dominated group among the cooking activities ($40.0 \sim 63.8\%$ of Σ PAHs) except cafeteria frying ($14.5 \pm 3.1\%$ of Σ PAHs). The six-ringed PAHs emitted from three conventional Chinese cooking ($21.2 \sim 31.0\%$ of Σ PAHs) were higher than those from two outdoor charbroiling ($11.1 \sim 17.6\%$ of Σ PAHs). For the seven-ringed PAHs, much higher contributions were seen for cafeteria frying and cafeteria boiling (avg. 12.4% of Σ PAHs), in comparison with only 2.2 ~ 4.9% of Σ PAHs for the rest of the cooking activities. It should be noticeable that five-ringed PAHs ($69.9 \sim 88.4\%$) were abundant for the two backgrounds, suggesting that besides certain amount of primary sources, secondary formation of PAHs from photolysis and/or oxidation by reacting with hydroxyl radicals, ozone, nitrogen oxides, or other strong oxidizing agents in the atmosphere should not be underestimated (Marr et al. 2005). Similarly, the highest contribution of five-ringed PAHs ($63.8 \pm 3.7\%$) for snack-street boiling could be explained by the combustion of materials and fuels at an open environment. In addition, four-, five-, and six-ringed PAHs totally accounted for 67.6 ~ 95.1% (avg. 87.5%) of Σ PAHs among the five cooking activities, consistent with the observations from a recent research conducted by Zhang et al. (2016).

PAHs are classified into three groups for further characterization: low molecular weight (LM-PAHs, with three aromatic rings), moderate molecular weight (MM-PAHs, with four

aromatic rings), and high molecular weight (HM-PAHs, with five to seven aromatic rings) (Chen et al. 2012). Figure 3 illustrates their distributions of the cooking activities samples. In this study, the percentage of LM-PAHs was 5.2% on average for all cooking activities, attributed to their high vapor pressure and most distribution in gas phase. HM-PAHs accounted for the largest proportion of Σ PAHs (avg. 69.2%). The two charbroiling and three conventional Chinese cooking had a contribution of 62.2 and 73.8% on average, respectively. Similar results were reported by Saito et al. (2014), which concluded that HM-PAHs were the most abundant proportion in the cooking exhaust. The HM-PAHs could not be completely decomposed or destroyed since those cooking temperatures were comparatively low. At a higher operation temperature such as in coal-fired power plants and heating/industrial boilers, the conditions facilitate for LM-PAH formation (Zhang et al. 2016). Considering that various sources of PAHs in the environments, it should be cautious to apportion the pollution origins using the ring number distribution.

Diagnostic ratios of PAHs

Diagnostic ratios of PAHs isomers serve as important tools for the identification of pollution sources (Tobiszewski and Namieśnik 2012; Yunker et al. 2002). Considering that PAHs may change from sources to receptors, it is better to use similar physico-chemical properties of PAH isomers to reduce the bias (Ravindra and Grieken 2008; Yao et al. 2015). In this study, > 50% of data were undetectable and/or below field blank for few PAHs (e.g., ACE, FLU, PHE, and ANT) (Table S2), their related ratios were excluded. As a result, five widely used diagnostic ratios, including BaA/(BaA + CHR), IPY/(IPY + BPE), BbF/BkF, FLT/(FLT + PYR), and BaP/(BaP + BPE), were investigated (Abdullahi et al. 2013; Shen et al. 2013; Yao et al. 2015).

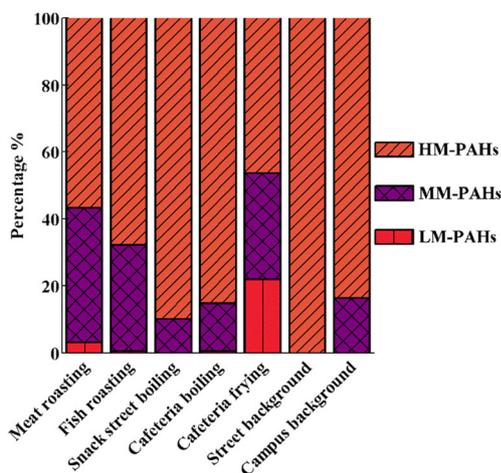


Fig. 3 Distribution of LM-PAHs, MM-PAHs, and HM-PAHs in the cooking activities and background samples

Table 3 compares the diagnostic ratios for the five cooking activities in this study and other sources in literatures. Close values were observed between the two charbroiling, and most of the ratios were similar among the three conventional Chinese cooking, except BbF/BkF. The BbF/BkF ratio for snack-street boiling was as high as 8.31, which was 5.9 and 7.1 times those for cafeteria boiling and frying, respectively. Such high value of BbF/BkF ratio could be ascribed to the coal combustion used as cooking fuel in snack street, which was also close to the value of 8.08 reported by Zhang et al. (2008).

The FLT/(FLT + PYR) ratios for meat roasting, fish roasting, and cafeteria frying ranged from 0.51 to 0.57, much higher than that of cafeteria boiling (0.37), ascribed to their large oil consumption. However, See and Balasubramanian (2008) found the FLT/(FLT + PYR) ratios for frying and boiling tofu were very similar. Other factors including fat contents, ingredients, and cooking fuels and temperatures could also contribute to the PAH emissions. In addition, FLT/(FLT + PYR) ratios for meat and fish roasting in this study were close to the charcoal combustion (0.49) (Sepetdjian et al. 2010), supporting the influences from cooking fuels.

On the other hand, slightly higher BaA/(BaA + CHR) ratios were found for the oil-based than water-based cooking activities in this study. The values were also high for cooking with Chinese oil (0.62) and rape seed oil (0.89) (Li et al. 2003; Zhu and Wang 2003). The ratio of BaA/(BaA + CHR) thus acts a good indicator for the influences of cooking oil.

For the ratios of IPY/(IPY + BPE) and BaP/(BaP + BPE), two charbroiling had much higher values (in the range of 0.39 ~ 0.68 and 0.39 ~ 0.69, respectively) in comparison with the three conventional Chinese cooking (in the range of 0.08 ~ 0.21 and 0.13 ~ 0.22, respectively). The values of IPY/(BPE + IPY) and BaP/(BaP + BPE) ratios were 0.55 and 0.53, respectively, for charcoal combustion (Sepetdjian et al. 2010), suggesting that these two diagnostic ratios efficiently appointed to the contributions of charcoal combustion.

In the background samples, the IPY/(IPY + BPE) and BaP/(BaP + BPE) ratios were close to the corresponding values of meat roasting, rice straw, and charcoal combustion, while the BbF/BkF ratios were similar with those of rice straw and diesel emission (Table 3). These observations prove that meat roasting was reasonably the largest contributor of PAHs, considering that charbroiling is one of the most popular dining in urban Yucheng. In addition, biomass burning and diesel exhaust also had significant impacts on atmospheric PAHs. Our interpretations with multiple diagnostic ratios could reduce bias to discriminate the potential pollution sources.

Toxicity evaluation

Figure 4 illustrates the concentrations of BaP, BaPE, and the calculated TEQ. The meat roasting had the highest

Table 3 Diagnostic ratios of PAHs from cooking activities and other sources

	FLT/(FLT + PYR)	IPY/(IPY + BPE)	BaA/(BaA + CHR)	BaP/(BaP + BPE)	BbF/BkF	Reference
Cooking activities						
Meat roasting	0.51	0.68	0.45	0.69	0.45	This study
Fish roasting	0.57	0.39	0.43	0.39	0.95	
Snack-street boiling	— ^a	0.13	0.31	0.18	8.31	
Cafeteria frying	0.52	0.08	0.46	0.13	1.41	
Cafeteria boiling	0.37	0.21	0.41	0.22	1.17	
Campus background	— ^a	0.53	0.18	0.60	1.30	
Street background	— ^a	0.59	— ^a	0.69	1.26	
Cooking in other studies						
Western cooking	0.46	0.63	0.38	0.71	0.93	Li et al. (2003)
Chinese cooking	0.50	0.63	0.62	0.43	1.07	
Boiling	0.52	0.52	0.30	0.23	1.89	See and Balasubramanian (2008)
Frying	0.54	0.77	0.27	0.45	0.64	
Charbroiling meat hamburger	0.32	0.38	0.23	0.44	0.78	Rogge et al. (1991), McDonald et al. (2003)
Seed oil	0.46	— ^b	0.89	— ^b	— ^b	Zhu and Wang (2003)
Other sources						
Coal	0.37	0.37	0.27	0.40	8.08	Zhang et al. (2008)
Rice straw	0.51	0.50	0.46	0.67	1.00	Sheesley et al. (2003)
Wood burning	0.43	0.19	0.21	0.56	0.91	Rogge et al. (1998)
Noncatalyst gasoline	0.61	0.04	0.57	0.23	0.93	Rogge et al. (1993)
Diesel	0.37	0.00	0.27	0.45	1.07	Rogge et al. (1993)
Charcoal	0.49	0.55	— ^b	0.53	— ^b	Sepetdjian et al. (2010)
Natural gas	0.51	— ^b	0.17	— ^b	0.72	Rogge et al. (1993)

^aNo ratio was calculated because > 50% of the data were undetectable and/or below field blank

^bBlank space means no data available

carcinogenic potencies shown by all three indexes. BaP is the sole PAH listed in the air quality standards in China with a daily average limit of 10 ng/m³ for ambient airs. The BaP concentrations of the five cooking activities were meat

roasting > cafeteria frying > fish roasting > cafeteria boiling > snack-street boiling in a descending order. It is obvious that the average concentrations of BaP for the three oil-based cooking activities were 1.2 ~ 2.6 times exceeded the Chinese ambient daily limit, while the values for the water-based cooking activities and backgrounds were well below the guideline. Hence, the significance of BaP emission from cooking activities should not be under-estimated, particularly for cooking involved large amounts of oils.

The trend of BaPE was similar with that of BaP, except higher value was seen for snack-street boiling than cafeteria boiling. The stall of snack-street boiling was next to the emissions from other frying and sautéing activities, which used coal as cooking fuel. Lower BaPE was also obtained for water-based than oil-based cooking activities, supporting that more formation of BaP, DBA, BbF, and BaA during oil thermal cooking (Chiang et al. 1999; Li et al. 1994).

For TEQ, the meat roasting had the highest value as well, followed by the snack-street boiling. The high TEQ for the snack-street boiling could be ascribed to the contribution of DBA (83.0% of TEQ), which had the highest TEF of 5.0 and

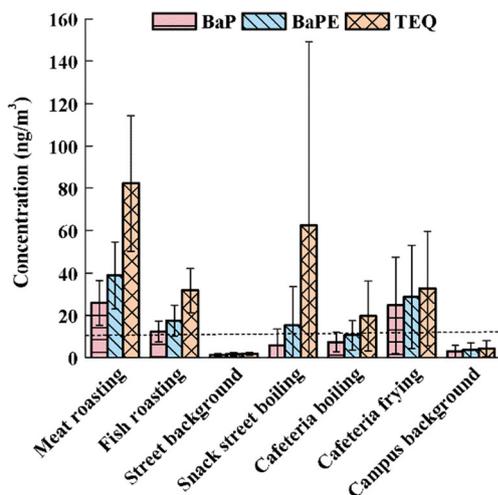


Fig. 4 Concentrations of BaP and calculated BaPE and TEQ

concentration of $15.0 \pm 21.5 \text{ ng/m}^3$ among the cooking activities.

The above three indexes merely refer to the toxicity of PAHs but cannot fully represent the toxicities of $\text{PM}_{2.5}$. Apart from PAHs, other organic and inorganic compounds (e.g., sulfate, nitrate, ammonium, and heavy metals) in $\text{PM}_{2.5}$ could also induce inflammation, oxidative stress, genetic toxicity, mutagenicity and carcinogenicity effects (Li et al. 2017; Wang et al. 2013; Zhang et al. 2017). Zhang et al. (2017) reported that the integrated risk of heavy metals was three orders of magnitude higher than those of PAHs from cooking sources, demonstrating that the toxicities for heavy metals should not be ignored. Furthermore, possible synergistic effect among PAHs and heavy metals might enhance the toxicity of $\text{PM}_{2.5}$ (e.g., damage the double helix structure of DNA), which is worth to be further investigated. Besides, nitrated and oxygenated PAHs could be formed from parent PAHs thru homogeneous and/or heterogeneous photo-oxidation reactions with atmospheric oxidants, photolysis, and thermal conversions. Some of these PAH derivatives are more toxic than parent PAHs (Alves et al. 2017). The toxicity of $\text{PM}_{2.5}$ from cooking source is probably magnified.

Conclusions

Both cooking method, fat and water contents in raw materials, oil consumption and cooking fuels could greatly contribute on the PAH formations in $\text{PM}_{2.5}$ from different cooking activities. Diagnostic ratios of isomer PAHs are useful tools to characterize the cooking sources. Meat roasting had the highest carcinogenic potencies indexed by the concentrations of BaP and values of BaPE and TEQ. In addition, oil-based cooking activities had higher BaP and BaPE indexes than water-based ones. Based on the results from this study, the significance of emission from roasting should not be overlooked. It is crucial to establish proper emission controls to regulate commercial roasting activities, which is one of the popular local dining in China. While the toxicity of $\text{PM}_{2.5}$ from cooking activities could be magnified by PAH derivatives and potential synergistic effects from other components in $\text{PM}_{2.5}$, further investigation works are needed.

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