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Characterization and health risk assessment of airborne pollutants in commercial restaurants in northwestern China: Under a low ventilation condition in wintertime



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HIGHLIGHTS

GRAPHICAL ABSTRACT

- Cooking activities impacted on dining areas of restaurants.
- Cooking methods and fuels are dominated sources.
- In-dining cooking and cigarette smoke contributed to indoor pollution.
- Cancer risk assessments demonstrate the environments were not optimistic.
- Reactive organic species were formed under poor ventilation environments.

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ABSTRACT

Impacts on indoor air quality of dining areas from cooking activities were investigated in eight categories of commercial restaurants including Szechwan Hotpot, Hunan, Shaanxi Noodle, Chinese Barbecue, Chinese Vegetarian, Korean Barbecue, Italian, and Indian, in Northwestern China during December 2011 to January 2012. Chemical characterization and health risk assessment for airborne carbonyls, and particulate-bound polycyclic aromatic hydrocarbons (PAHs) and heavy metals were conducted under low ventilation conditions in wintertime. The highest total quantified carbonyls ($\Sigma_{carbonyls}$) concentration of 313.6 µg m⁻³ was found in the Chinese Barbecue, followed by the Szechwan Hotpot (222.6 µg m⁻³) and Indian (221.9 µg m⁻³) restaurants. However, the highest $\Sigma_{carbonyls}$ per capita was found at the Indian restaurant (4500 µg capita⁻¹), suggesting that cooking methods such as stir-fly and bake for spices ingredients released more carbonyls from thermal cooking processes. Formaldehyde, acetaldehyde, and acetone were the three most abundant species, totally accounting for >60% of mass

airborne carbonyls

Hydroxyl radic

PAHs heavy metals

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Heavy metals Commercial restaurants Cancer risk assessment concentrations of the $\Sigma_{carbonyls}$. Phenanthrene, chrysene, and benzo[*a*]anthracene were the three most abundant PAHs. Low molecular weight fraction (Σ PAHs_{S178}) had the highest contributions accounting for 40.6%–65.7%, much greater than their heaver counterparts. Diagnostic PAHs ratios suggest that cooking fuel and environmental tobacco smoke (ETS) contribute to the indoor PAHs profiles. Lead was the most abundant heavy metal in all sampled restaurants. High quantity of nickel was also found in samples due to the emissions from stainless-steel made kitchen utensils and cookware and ETS. Cancer risk assessments on the toxic substances demonstrate that the working environment of dining areas were hazard to health. Formation of reactive organic species (ROS) from the cooking activities was evidenced by measurement of hydroxyl radical (•OH) formed from simulating particulate matter (PM) react with surrogate lung fluid. The highest •OH concentration of 294.4 ng m⁻³ was detected in Chinese Barbecue. In addition, the elevation of the concentrations of PM and •OH after non-dining periods implies that the significance of formation of oxidizing-active species indoor at poor ventilation environments.

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1. Introduction

Fuel combustions for cooking and heating are one of the major sources of indoor air pollution which influence half of population in the world (WHO, 2002). Cooking methods such as grill, deep and stir fries, bake, and boil can produce many organic and inorganic air pollutants. Foods, ingredients, and cooking oils as raw materials produces different emissions from cooking processes. High operation temperatures may lead to different degrees of degradation of nutrients (i.e., sugar, protein, and fat), which can form a series of primary pollutants in cooking oil fumes (COFs) (Shields et al., 1995; Lee et al., 2001; Svendsen et al., 2002; Pan et al., 2008; Kabir and Kim, 2011; Alves et al., 2014).

Airborne carbonyls, polycyclic aromatic hydrocarbons (PAHs), heavy metals, and particulate matter (PM) are often observed in cooking emissions (Shields et al., 1995; Lee et al., 2001; Svendsen et al., 2002; Lund and Petersen, 2006; Kabir and Kim, 2011). Many of these species are known carcinogens (IARC, 2016) (Table 1). Formaldehyde can cause nasopharyngeal cancer (IARC, 2004) and is suggested to be potentially associated with leukemia (Zhang et al., 2009a, 2009b) and acetaldehyde is also a suspected carcinogen (Baez et al., 2003; Zhang et al., 1994). Zhang et al. (2009a, 2009b) reported that as much as 1.6% of lung cancer cases in China can be ascribed to inhalation of PAHs in polluted air. Besides, anthropogenic fine particules are often enriched with toxic heavy metals, which are of particularly concern on human health (Dockery and Pope, 1996). Many epidemiological studies

Table 1

Classification of carcinogenic carbonyls, PAHs and heavy metals (IARC, 2016).

Compounds	Classification ^a
Formaldehyde Acetaldehyde Fluorene	Group 1, 2012 Group 2B, 1999 Group 3, 2010
Phenanthrene	Group 3, 2010
Benzo[b]fluoranthene	Group 2B, 2010
Benzo[k]fluoranthene	Group 2B, 2010
Benzo[a]pyrene	Group 1, 2012
Indeno[1,2,3-cd]pyrene	Group 2B, 2010
Benzo[ghi]pyrene	Group 3, 2010
Dibenzo[<i>a</i> , <i>h</i>]anthracene	Group 2A, 2010
Chromium (Cr)	Group 3, 1990
Cobalt (Co)	Group 2B, 1991
Nickel (Ni)	Group 2B, 1990
Arsenic (As)	Group 1, 2012
Selenium (Se)	Group 3, 1987
Cadmium (Cd)	Group 1, 2012
Mercury (Hg)	Group 3, 1993
Lead (Pb)	Group 2B, 1987

Group 2A: Probably carcinogenic to humans.

Group 2B: Possibly carcinogenic to humans.

Group 3: Not classifiable as to its carcinogenicity to humans.

^a Group 1: Carcinogenic to humans.

demonstrated that inhalation of the toxic pollutants from cooking can raise lung cancer risks, obviously even in non-smokers (Seow et al., 1998; Seow et al., 2000; Parkin et al., 2005; Samet et al., 2009; Yuan et al., 2014). In addition, Asian women have a higher incidence of lung cancer than men despite their smoking prevalence is comparatively lower (Seow et al., 2000). Cooking emission pollutants can be thus considered as potential carcinogens. The adverse health effects from ambient PM are linked to the formation of reactive oxygen species (ROS) in cardiopulmonary tissues. While hydroxyl radical (•OH) is the most reactive species, it can react with different organic molecules at diffusioncontrolled rate constants (Held et al., 1996; Forman et al., 2010).

Xi'an (33.42°N-34.45°N and 107.40°E-109.49°E), the capital city in Northwestern China, has a population of over 8 million in an urban area of 1066 km². There were >1000 food production enterprises and 24,757 catering service licensed restaurants in 2015 (Zhao et al., 2016). However, indoor air quality of dining areas impacted by different cooking activities is often overlooked. Currently, researches are mostly focused on emission profiles from cooking processes, while air samples were collected directly from the exhausts in kitchens. According to the Chinese National Data in 2014, the total number of restaurant employees was 2.5 million in Mainland of China (National Bureau of Statistics of China, 2014). The health of the workers who serve in dining areas (e.g., waiters, waitress, and cashiers) must be taken into account. This is particularly critical due to low indoor ventilation rates resulted from heat preservation in northern China in winter. In this study, air samples were collected in dining areas of common types of restaurants at an urban center in Xi'an city. Chemical profiles were obtained and factors affected the indoor air quality of dining places were determined. Health assessments were also conducted regarding to pollutant levels. The results are essential reference for establishment of local regulations for indoor air quality in commercial restaurants.

2. Data and methods

2.1. Sampling site description

Eight commercial restaurants were selected in this study based on their ubiquity. They were all located in the Great Wild Goose Pagoda at Qujiang District in southeastern Xi'an, and catalogized as Szechwan Hotpot (R1), Hunan (R2), Shaanxi Noodle (R3), Chinese Barbecue (R4), Chinese Vegetarian (R5), Korean Barbecue (R6), Italian (R7) and Indian (R8). Table 2 lists the details of each restaurant, including major foods served, ingredients involved, cooking methods, fuel types, exhausts in kitchens, and ventilation systems equipped in dining areas.

All ground-floor restaurants were selected in this study. The kitchen was located at the same level which separated from the dining area in each restaurant. The kitchen door was kept closure all the times, and most dishes were transferred through a pick-up window ($<0.5 \text{ m}^2$) on the wall of kitchen. As the sampling campaigns were conducted in winter, heat was supplied to indoors through central hot-water circulation

A summary of characteristics of sampled commercial restaurants.

Restaurants	Sites	Major food served	Cooking methods	Cooking implements	Number of customers	Number of smokers	T (°C)	RH (%)	Area (m ²)	Type of air-conditioner	In-dining cooking activities	Fuel type	Exhaust system
Szechwan hotpot	R1	Various; red peppers	Boil	Metal pot	65	0	11.7	46.5	85	Central	Ν	Electricity, alcohol	Electrostatic precipitator
Hunan	R2	Various; green/red peppers	Stir-fry, steam, boil	Iron wok	715	4	17.1	38.0	140	Packaged	Ν	Natural gas	Hydrovent
Shaanxi noodle	R3	Pork; vegetable; noodles; marinated meat in baked bun	Boil, steam, stir-fry, bake	Iron wok	430	25	16.6	34.6	1000	Central	Y	Natural gas, coal ^a	Hydrovent
Chinese barbecue	R4	Meats (pork, beef, mutton, chicken), fish, vegetable	Grill	Charbroiled	165	12	16.5	36.1	180	Packaged	Ν	Natural gas	Electrostatic precipitator
Chinese vegetarian	R5	Tofu; vegetable; bread	Stir-fry, boil, steam	Iron wok	252	0	18.6	32.6	400	Central	Ν	Natural gas	Hydrovent
Korean barbecue	R6	Meat; fish; vegetable	Grill	Metal tray/metal mash	240	0	25.2	25.1	150	Central	Y	Electricity, alcohol	Electrostatic precipitator
Italian	R7	Beef, chicken, hamburger, potato chip	Pan-fry, roast, bake	Iron plate/fry pan	156	0	17.1	32.2	500	Central	Ν	Natural gas	Hydrovent
Indian	R8	Beef, chicken, curry; potato	Boil, stir-fry	Metal pot	409	0	19.1	41.4	150	Central	Ν	Natural gas	Hydrovent

^a Coal was used for boiling water in dining area.

systems. In addition, all windows in dining areas were closed and the entrances of restaurants were covered with thick curtains to avoid any loss of heat energy to outdoors. The dining areas were on a range of 80 to 1000 m². To minimize the inconsistency, no decoration had been done and no new furnishing had been installed since their businesses started synchronously between the years of 2009-2010. Indoor smoking was not forbidden which was found in Hunan, Shaanxi Noodle and Chinese Barbecue. The number of smokers were recorded but the quantities and classes of cigarettes could not be counted. Among the eight sampled restaurants, in-dining cooking was found in the Szechwan Hotpot and Korean Barbecue where most of the foods were cooked by diners. A coal-burning stove for water heating was located in the dining area of Shaanxi Noodle. Central ventilation systems were operated in each restaurant except Hunan and Chinese Barbecue, where packaged air conditioners were used. An individual exhaust head was installed over each dining table in the Korean Barbecue to withdraw the cooking flames. The ventilation rate, as expressed in term of air changes per hour (ACH), was evaluated with the changes of carbon dioxide (CO₂) concentration in dining area (Clements-Croome et al., 2008). In this study, the ACH was 0.11 to 0.75 h^{-1} in unit per hour, demonstrating a poor infiltration and exchange of airs in the indoor air (Clements-Croome et al., 2008; Xu et al., 2015). For outdoor, no obvious pollution sources were found nearby. The restaurants were at least 100 m far away from the roadside.

2.2. Sampling collection

Air sampling campaign was conducted from December 2011 to January 2012. To ensure the representativeness of the data, gaseous and PM samples were collected in two dining periods (12:00–14:00 and 18:30–20:30 LST for lunch and supper hours, respectively) synchronously. Baseline samples (non-dining period) were taken during break period (15:00–17:00 LST) while no diner was present and cooking activities were terminated. Sampling equipment was installed in the center of indoor dining places and 1.5 m above the ground level at each sampling site. Airborne carbonyls were collected into silica cartridges impregnated with acidified 2,4-dinitrophenylhydrazine (DNPH) (Sep-Pak DNPH-silica, 5 µm particle size, 125 Å pore size; Waters Corporation, Milford, MA) following the instructions on the compendium method TO-11A established by the United States Environmental Protection Agency (U.S.EPA) (U.S.EPA, 1999). Each cartridge sample was collected at a flow rate of 0.7 L min⁻¹ for 2 h. PM_{2.5} samples were collected on pre-fired (780 °C, 3 h) 47 mm quartz-fiber filters (QM/A, Whatman, Clifton, NJ, USA) with mini-volume air samplers (Airmetrics, Eugene, OR, USA) which operated at a flow rate of 5 L min⁻¹. No PM samples were taken at Chinese Vegetarian and Italian restaurants due to sampler faults. All cartridges and filter samples were stored in a refrigerator at <4 °C prior to chemical analysis. Indoor temperature and relative humidity (RH) were recorded during the sampling periods.

2.3. Airborne carbonyls analysis

Twenty target carbonyls (including mono- and di carbonyls) in the cartridges were quantified. Each cartridge was eluted with 2 mL acetone-free acetonitrile (ACN; HPLC grade, J&K Scientific Ltd., Ontario, Canada) into an amber volumetric vial. Twenty milliliters of the sample were injected to an Agilent Technology high-pressure liquid chromatography (HPLC) coupled with a photodiode array detector (DAD) (Series 1200; Santa Clara, CA, USA). The analytes separation was conducted with a PerkinElmer Spheri-5 ODS 5 µm C-18 reversed-phase column $(4.6 \times 250 \text{ mm}; \text{Norwalk}, \text{CT}, \text{USA})$, which maintained at 30 °C. The mobile phase system involves: mixture A, 6:3:1 (v/v) of water/ acetonitrile/ tetrahydrofuran; mixture B, 4:6 (v/v) of water/acetonitrile and mixture C, acetonitrile. The gradient program was A: B (80:20, v/v) for 1 min, followed by linear gradients of A:B (50:50, v/v) for the next 8 min, then B (100%) in 10 min, and C (100%) in 6 min, and finally kept for 5 min. A gradient flow rate of 2 mL min⁻¹. The wavelengths of 360, 390 and 420 nm were applied for identifying aliphatic, aromatic, and di carbonyls, respectively. Certified calibration standards of mono carbonyl DNP-hydrazones were purchased from Supelco (Bellefonte, PA) and diluted to a range of $0.015-3.0 \text{ mg mL}^{-1}$. Di carbonyl DNP-hydrazones were prepared by mixing known amounts of glyoxal and methylglyoxal from Sigma (St. Louis, MO, USA) in acetonitrile with 10 folds of excess DNPH in an acidic aqueous solution. The mixtures were stood at room temperature for at least 6 h for a complete derivatization (Dai et al., 2012). The calibration standards for the di carbonyl DNP-hydrazones were then prepared into a range of 0.01 to 2.0 μ g mL⁻¹. Linearity of each calibration was presented by a correlation of determination (r^2) of at least 0.999. The minimum detection limit (MDL) was estimated by analyzing a minimum of seven replicates of a standard solution containing the analytes at the lowest concentration of 0.015 mg mL⁻¹. The MDLs of the target carbonyls ranged from

0.002 to 0.010 mg mL⁻¹ which can be translated to 0.016–0.12 ppbv with a sampling volume of 2.02 m³ in this study. The measurement precisions are in a range of 0.5–3.2% for the quantified carbonyls (Ho et al., 2015).

2.4. PM measurements

2.4.1. Gravimetric method

The PM_{2.5} samples were weighed with a Sartorius ME 5-F electronic microbalance (Sartorius, Gottingen, Germany) at a sensitivity of $\pm 1 \mu g$. The filters were equilibrated at temperature of 20–23 °C and RH of 35–45% inside a conditioned chamber for 24 h, and weight at least two times prior and after the sampling events. The allowable absolute errors between duplicate weights were set to $\leq 0.015 \text{ mg}$ and ≤ 0.020 for blank filters and loaded samples, respectively.

2.4.2. Organic analysis

Particulate-bound 13 PAHs and *n*-alkanes $(C_{16}-C_{36})$ were quantified with in-injection port thermal desorption (TD)-gas chromatography/ mass spectrometry (TD-GC/MS) method. Detail experimental procedures were described in our previous publications (Ho and Yu, 2004; Ho et al., 2008, 2011). Briefly, aliquots of the filters $(0.53-2.63 \text{ cm}^2)$ were cut into small pieces and inserted into a TD tube which has the same dimensions as the GC/MS injector liner. The injector port was lowered to 50 °C before analysis and then raised to 275 °C for desorption in a splitless mode while the GC oven temperature was kept at 30 °C. After the injector temperature reached the set point, the oven program started. The MSD was scanned from 50 to 550 amu under electron impact ionization (EI) at a voltage 70 eV and source temperature of 230 °C. Identification was achieved by characteristic ion and comparing the retention times of the chromatographic peaks with those of standards. The non-dining period filters were analyzed using these same procedures. A five-point calibration over a concentration range of 1-10 ng for each PAH (Sigma-Aldrich, Bellefonte, PA, USA) from a standard mixture was established. The correlation coefficients (R²) for linear regressions of the calibration curves were >0.99. For each ten samples, one replicate analysis was done; the relative standard deviation of these replicates was from 1.7 to 14.8% for PAHs. All data were corrected for the average value of the blanks.

2.4.3. Elemental analysis

Eight elements on the filter samples were quantified using an Energy Dispersive X-Ray Fluorescence (ED-XRF) spectrometry (Epsilon 5, PANalytical, B.V., Netherlands). The X-ray is from a side window X-ray tube with a gadolinium anode. The characteristic X-ray radiation is detected by a germanium detector (PAN 32) and 30 min are required for each sample to obtain a spectrum of X-ray counts versus photon energy, with the individual peak energies matching specific elements, and peak areas corresponding to elemental concentrations (Brouwer, 2003). The ED-XRF spectrometer was calibrated with thin-film standards obtained from MicroMatter Co. (Arlington, WA, USA). Replicate analysis were conducted on every eighth sample. The analytical precisions were obtained from five replicate analysis of one quartz-fiber filter sample which ranged from 5.1-22.2%, which met the quality assurance objectives for the target carcinogenic heavy metals. Laboratory blanks of the quartz-fiber filters were analyzed to derive a MDL of 2-21 ng m⁻³. National Institute of Standards and Technology (NIST) Standard Reference Material (SRM) 2783 was employed to validate the accuracy of measurement. The relative errors of 0.6%, 0.16%, 0.20%, 0.24%, 0.08%, 0.62%, 0.18% and 0.14%, respectively, were reported for Pb, Co, Ni, As, Se, Cd, Hg and Cr (Abbreviation are shown in Table 1). No analytical data for the samples of Szechwan Hotpot and Shaanxi Noodle due to insufficient filter for measurement.

2.4.4. Hydroxyl radical (•OH) analysis

ROS formed from inhaled PM in cardiopulmonary tissues is critical to access the oxidative damage to human health (Dockery et al., 1993; Pope III et al., 1995; Pope et al., 2004; Pekkanen et al., 2002; Pope and Dockery, 2006). While •OH is the most reactive form among those ROS species, quantitation of its potential formation concentration was conducted as the same approach shown in Shen and Anastasio (2011) and Jung et al. (2006).

The analysis involved simulating the PMs react with surrogate lung fluid when they entered human's lung. In brief, a punch of filter (1.5 to 2.0 cm²) was placed in a 10-ml Teflon tube. Six milliliters of surrogate lung fluid were added into the tube, with a mix of ascorbic acid at a final concentration of 50 µM. The vial was thoroughly wrapped with aluminum foil, placed on a wrist-action shake table, and shaken in the dark at room temperature for 24 h. The concentration of •OH was measured using 10 mM benzoate as a chemical probe (Anastasio and McGregor, 2001). A 500 µL aliquot of PM extraction solution was added into phydroxybenzoic acid (p-HBA). The extract was guickly filtered using a 0.22 µm syringe filter and transferred into an autosampler vial, where 100 µM desferoxamine mesylate (DSF) and 50 µM bisulfite ion (HSO_3^-) were added to guench •OH generation. After 10 min, the extract was acidified to pH 2 by adding 5 µL of 1.0 M sulfuric acid (H₂SO₄). The final solution was injected into an Agilent 1200 HPLC/DAD system (Palo Alto, CA, USA) and the absorbance of product was detected at a wavelength of 254 nm.

2.5. Cancer risk assessment method

The carcinogenicity of formaldehyde, acetaldehyde, and particlebounded PAHs and heavy metals are confirmed by World Health Organization (WHO) and are listed in the carcinogenic lists (IARC, 2016). The method of calculating cancer risk is used according to U.S.EPA (US EPA, 1989, 1992, 2004; Cavalcante et al., 2006). The equation for cancer risk assessment is:

$$CR = CDI * CSF_{inh}$$
(1)

where CDI (chronic daily intake) expresses the intake of carcinogenic pollutants per day by adult and CSF illustrates the slope factor when the risk gradient factor of toxicity of carcinogenic pollutants in unit inhalation is considered as liner.

CSF is an upper-bound estimate of increased cancer risk from a lifetime exposure to an agent (US EPA, 1997, 2001). It is used to illustrate the unit proportion affected by a certain contamination per mg of agent per kg of body weight per day (US EPA, 2001a). CSF is calculated by the following equation:

$$CSF_{inh} = URF_{inh} * BW * 1000/IR$$
⁽²⁾

where URF is inhalation unit risk factor (μ g/m³), which is the upperbound excess lifetime cancer risk from continuous exposure to an agent at a concentration of 1 μ g m⁻³ in air (US EPA, 2001a). According to U.S.EPA, the URF of formaldehyde is ($1.3 \times 10^{-5} \mu$ g m⁻³)⁻¹ and acetaldehyde is $2.2 \times 10^{-6} (\mu$ g m⁻³)⁻¹ (US EPA, 1989); BW is the body weight (kg) of an individual which is 66.2 kg (male) and 57.3 kg (female) (National Health and Family Planning Commission, 2015); 1000 is unit conversion (μ g m⁻³) and IR is inhalation rate (m³ day⁻¹) which is 20 m³ day⁻¹.

The equation for calculation of CDI is:

$$CDI = C * I_{ra} * ED * EF * L * (BW * ATL * NY)^{-1}$$
(3)

where C (mg/m³) is the concentration of the pollutant; I_{ra} is inhalation rate (m³ hr⁻¹) which is 0.83 (US EPA, 1997); ED is exposure time (hr week⁻¹) which is 48 h per week; L is the length of exposure (years) and we assumed that the working age is from 18 to 60 (42 years); EF is the exposure frequency (52 week year⁻¹, except Spring

Carbonyl concentrations (µg/m³) in the exhaust of sampled commercial restaurants.

Concentration	Szechwan hotpot		Hunan		Shaanxi noodle		Chinese barbecue		Chinese vegetarian		Korean barbecue		Italian		Indian	
	Lunch	Supper	Lunch	Supper	Lunch	Supper	Lunch	Supper	Lunch	Supper	Lunch	Supper	Lunch	Supper	Lunch	Supper
Formaldehyde (C1)	11.0	12.2	19.4	23.0	14.5	15.9	_ ^a	66.5	21.4	20.0	30.1	34.3	17.0	11.8	27.4	30.1
Acetaldehyde (C2)	19.2	78.3	21.8	35.4	24.6	25.8	-	48.7	23.6	28.4	27.5	34.9	19.3	15.9	47.9	50.4
Acetone	20.0	46.6	23.1	30.8	20.1	19.2	-	25.1	22.8	23.7	26.4	30.3	18.1	13.8	45.8	54.3
Propionaldehyde (C3)	4.0	7.4	4.4	4.3	4.0	2.9	-	7.6	3.9	4.5	4.5	5.4	3.6	2.5	8.3	8.8
Methyl ethyl ketone (MEK)	2.8	9.4	3.5	6.7	4.8	4.4	-	6.7	5.8	6.4	6.9	7.3	4.7	3.7	10.1	8.7
iso- + n-Butyraldehyde (iso + nC4)	1.1	3.6	1.2	1.9	1.7	1.3	-	3.4	1.4	1.6	1.9	2.2	1.2	0.8	2.7	2.6
Benzaldehyde (benz)	2.0	3.6	1.9	2.3	3.1	2.9	-	3.7	2.7	2.7	2.4	2.8	2.2	1.3	4.1	3.8
iso-Valeraldehyde (iso-C5)	2.1	9.7	3.3	5.3	3.4	2.8	-	4.9	3.1	3.7	4.2	4.1	2.8	2.3	5.9	5.6
n-Valeraldehyde (nC5)	5.0	3.4	3.6	2.7	3.1	2.2	-	7.0	1.9	2.0	2.1	2.2	1.8	1.4	4.2	4.4
o-Tolualdehyde (o-tol)	bd ^b	bd	bd	bd	bd	bd	-	bd	bd	bd	bd	bd	bd	bd	bd	bd
m-Tolualdehyde (m-tol)	0.7	0.9	0.4	0.5	0.9	0.7	-	1.2	0.5	0.5	0.9	0.9	0.4	0.1	1.2	0.8
p-Tolualdehyde (p-tol)	2.6	2.0	0.5	0.7	0.1	0.1	-	3.1	0.2	0.1	0.8	0.5	0.1	0.1	0.2	0.4
Hexaldehyde (C6)	23.3	12.6	24.6	8.0	11.1	6.9	-	16.3	7.6	6.8	7.1	7.6	7.6	4.6	13.5	19.1
2,5-Dimethylbenzaldehyde (2,5-dB)	0.7	1.5	1.1	1.4	0.6	0.5	-	1.5	1.6	0.9	0.9	1.4	1.1	0.6	1.0	1.0
Heptaldehyde (C7)	7.3	10.1	3.2	4.5	4.5	3.0	-	63.7	2.7	2.6	4.5	6.6	2.4	1.7	8.4	11.0
Octaldehyde (C8)	3.3	4.2	2.6	2.3	3.6	2.7	-	16.2	2.5	2.3	3.5	4.0	2.2	1.4	4.3	5.2
Nonaldehyde (C9)	33.0	8.6	5.1	5.3	7.3	5.1	-	26.8	5.7	4.6	8.0	7.8	4.4	3.2	8.7	10.9
Glyoxal (gly)	3.4	5.3	2.3	3.8	1.3	1.5	-	5.3	5.7	3.4	2.3	2.2	2.7	1.7	2.2	2.2
Methylglyoxal (mgly)	1.3	3.4	2.1	3.6	1.7	1.4	-	5.8	3.4	2.0	2.2	1.9	1.8	1.1	2.5	2.5
Total carbonyls	142.9	222.6	124.3	142.6	110.3	99.5	-	313.6	116.4	116.0	136.4	156.3	93.5	68.0	198.3	221.9
Weight per capita (µg capita ⁻¹)	1020	1105	630	385	4000	4500	-	1530	1900	2100	4275	1762.5	6500	2000	7425	1575
Average	1063		490		4250		1530		2000		3038		4250		4500	

^a No sample was collected without any cooking activity.

^b Below detection limit.

Festival vacation); BW is the body weight (kg) of an adult which is 66.2 kg (male) and 57.3 kg (female) in China; ATL is the average time of life (70 years) and NY is the number of days per year (365 days).

3. Results and discussion

3.1. Presenting pollutants and background levels

Pollutant levels and compositions in different restaurants can vary with dining area sizes and designs, efficiencies of ventilation systems, foods and ingredients, cooking fuels and methods, and exhaust system in kitchens. All restaurants were being operated as usual during the sampling events. No obvious variations in either absolute values or molar compositions, statistically the same by Student's *t*-test with a 95% confidence level, were observed on the samples collected at each restaurant from the three visits. The average value was thus taken to demonstrate characteristics of different categories of restaurants. The significance of cooking emissions to the dining area airs was demonstrated by elevations of indoor pollutants levels. Therefore, data of airborne carbonyls, PAHs, and heavy metals reported afterward has been

subtracted by the average of corresponding baselines collected during the non-operating hours.

3.2. Characterization of airborne carbonyls

The concentrations of target carbonyls measured in two dining periods are listed in Table 3. The highest total quantified carbonyls ($\Sigma_{carbonyls}$) concentration of 313.6 µg m⁻³ was seen in the Chinese Barbecue, followed by Szechwan Hotpot (222.6 µg m⁻³) and Indian (221.9 µg m⁻³) restaurants. In comparison, much lower concentrations were found in Italian, Shaanxi Noodle, and Chinese Vegetarian restaurants. The concentrations were subjected to the numbers of diners, so there is no obvious pattern for lunch and supper among the restaurants. The concentrations of formaldehyde measured in all dining areas were all below the guideline level of 100 µg m⁻³ established by Standard Press of China (GB 3095–2012).

Three critical parameters, (i) the quantity of foods, (ii) efficiency of ventilation system, and (iii) the volume of restaurants, must be considered in order to compare the influences of each cooking activity on air quality of dining areas, instead of presenting their absolute concentrations. In this study, no statistical data on the quantity of foods and



Fig. 1. The average airborne carbonyl compositions in the samples collected in the restaurants.

Concentrations of PAHs and carcinogenic heavy metals in sampled commercial restaurants (ng m⁻³).

Compounds	Szechwan Hotpot		Hunan		Shaanxi Noodle		Chinese Barbecue		Korean Barbecue		Indian	
	ng/m ³	ng capita ⁻¹										
Fluorene (Group 3, 2010)	7.5	44.1	100.0	352.3	220.0	8855.3	143.0	697.6	205.0	3400.0	274.0	3269.2
Phenanthrene (Group 3, 2010)	96.7	566.9	335.0	1180.3	878.0	35,340.5	419.0	2044.1	712.0	11,808.9	875.0	10,439.9
Anthracene	30.2	177.0	54.5	192.0	138.0	5554.7	59.9	292.2	111.0	1841.0	177.0	2111.8
Fluoranthene	14.8	86.8	27.2	95.8	87.6	3526.0	42.0	204.9	62.8	1041.6	88.2	1052.3
Pyrene	15.3	89.7	26.8	94.4	86.7	3489.8	48.6	237.1	56.3	933.8	80.0	954.5
Benzo[a]anthracene	15.3	89.7	31.7	111.7	283.0	11,391.1	69.9	341.0	36.4	603.7	70.3	838.8
Chrysene	23.9	140.1	51.7	182.2	315.0	12,679.1	114.0	556.2	83.0	1376.6	128.0	1527.2
Benzo[b]fluoranthene (Group 2B, 2010)	17.3	101.4	35.8	126.1	234.0	9418.8	47.1	229.8	56.8	942.1	90.9	1084.6
Benzo[k]fluoranthene (Group 2B, 2010)	11.7	68.6	27.2	95.8	96.0	3864.1	22.2	108.3	40.3	668.4	49.9	595.4
Benzo[<i>a</i>]pyrene (Group 1, 2012)	21.3	124.9	49.7	175.1	247.0	9942.0	35.1	171.2	61.8	1025.0	99.3	1184.8
Indeno[1,2,3-cd]pyrene (Group 2B, 2010)	25.4	148.9	52.7	185.7	273.0	10,988.6	36.1	176.1	80.7	1338.5	119.0	1419.8
benzo[ghi]perylene (Group 3, 2010)	14.1	82.7	27.5	96.9	148.0	5957.2	23.0	112.2	44.6	739.7	54.4	649.1
Dibenzo[<i>a</i> , <i>h</i>]anthracene (Group 2A, 2010)	6.1	35.7	8.5	29.9	38.9	1565.8	12.9	62.9	15.0	248.8	18.2	217.1
Cr(Group3, 1990)	_ ^a	-	246.0	866.7	-	-	350.0	1707.5	208.0	3449.8	151.0	1801.6
Co(Group2B, 1991)	-	-	bd ^b	bd	-	-	104.0	507.4	95.0	1575.6	9.0	107.4
Ni(Group2B, 1990)	-	-	227.0	799.8	-	-	189.0	922.1	123.0	2040.0	104.0	1240.9
As(Group1, 2012)	-	-	bd	bd	-	-	bd	bd	bd	bd	bd	bd
Se(Group3, 1987)	-	-	bd	bd	-	-	bd	bd	bd	bd	bd	bd
Cd(Group1, 2012)	-	-	bd	bd	-	-	bd	bd	bd	bd	76.0	906.8
Hg(Group3, 1993)	-	-	662.0	2332.4	-	-	605.0	2951.5	529.0	8773.8	652.0	7779.2
Pb(Group2B, 1987)	-	-	2174.0	7659.6	-	-	1966.0	9591.3	1720.0	28,527.2	1909.0	22,776.8

^a No data for heavy metal was collected due to insufficient filters for chemical analysis.

^b Below detection limit.

ingredients were obtained due to a lack of information from the restaurants owners. An assumption had been thus made that each diner had the same quantity of foods in each restaurant. After taken into account of their individual air exchange rate and volume, we hence reported that the data as $\Sigma_{carbonyls}$ per capita (µg capita⁻¹) for the comparison.

The highest $\Sigma_{carbonyls}$ per capita was observed at the Indian restaurant (4500 μ g capita⁻¹), followed by Italian and Shaanxi noodle restaurants (both of 4250 μ g capita⁻¹). The major cooking methods applied in Indian restaurant include stir-fly, bake, and boil, and their ingredients typically contain strong-smelling spices, such as chili pepper and curry. These ingredients and spices will release irritant gas during the high temperature cooking processes (Feng et al., 2008). Baking of breads could also generate high amounts of incomplete combustion organic compounds. For the Italian restaurant, meats were cooked through pan-fly and potatoes were baked or deep-flied. These cooking methods require a large quantity of high-temperature oils (Siao and Rajasekhar, 2008). High value was also found for Korean Barbecue where the diners grilled the meats and small amount of vegetables (i.e., mushrooms and potatoes) with oils over the hot metal-plates on the dining tables. Even through an individual extractor was installed on the top of each table, the emissions from in-dining cooking could still influence the indoor airs (Ho et al., 2012). In comparison, another in-dining cooking restaurant of the Szechwan Hotpot had much lower $\Sigma_{carbonvls}$ per capita. Most of the foods were boiled in soups containing Chinese pepper but no any heating oil was applied. Our observations were thus consistent with the expectation that the boiling would not lead the formation of large amounts of carbonyls from foods and ingredients at a comparatively low temperature (~100 °C). It should be noted that even though boiling was the major cooking method for the Shaanxi Noodle restaurant, where a high $\Sigma_{carbonyls}$ per capita was calculated. The stove for boiling hot water for diner could be a source of incomplete combustion products as it used coal as a fuel. The lowest value (490 µg capita⁻¹) was detected for the Hunan restaurant where stir-fly is the dominated cooking method.

Fig. 1 demonstrates the carbonyl compositions of the indoor airs. Formaldehyde, acetaldehyde, and acetone are the three most abundant species, totally accounting for >60% of the $\Sigma_{carbonyls}$ on basis of mass concentrations. The highest contribution of formaldehyde was seen in Chinese Barbecue (40.8%), followed by Korean Barbecue (37.1%), Chinese

Vegetarian (31.1%), and Italian 31.0%. The results reveal that the cooking methods of grill and deep-fly could produce higher proportions of formaldehyde, in comparison with pan- and stir-fries, boil, and steam (Siao and Rajasekhar, 2008). The similarity is that both grill and deep-fly consume a large quantity of oils. Acetaldehyde is the most dominant carbonyl in the airs collected in Szechwan Hotpot (37.0%), and Shaanxi Noodle (30.4%) and Indian (28.8%) restaurants where boiling was sole or the major cooking method. Acetone is the third abundant carbonyls (~20% of the $\Sigma_{carbonyls}$) in all sampled restaurants, except Chinese Barbecue with the lowest of 7.9%. Larger compositions of long-chain carbonyls ($\Sigma_{carbonyls}$ (n=6.7.8.9) were seen on Chinese Barbecue and Szechwan Hotpot, which had a contribution of 19.0% and 14.4%, respectively, than those in other restaurants (~7%). For these two restaurants, it is an obvious smell of smoke aroma from a mix of condiments in the dining areas. It is no doubt that the foods and ingredients could participate in the carbonyls formation. For dicarbonyls, glyoxal and methylglyoxal were detectable in all sampled restaurants. The highest contribution of glyoxal was seen in Chinese Vegetarian (3.4%), with a contribution of foods and cooking style, but no trend was observed.



Fig. 2. The average concentrations and distributions of PAHs in the six restaurants.



Fig. 3. Average compositions of target carcinogenic heavy metals in four sampled restaurants.

3.3. Particulate-bound PAHs

The concentrations of 13 target PAHs are shown in Table 4. Shaanxi Noodle had the highest total quantified PAH concentrations (Σ PAHs) of 3045.2 ng m⁻³ among the sampled restaurants, following by Indian (2124.2 ng m^{-3}) and Korean Barbecue $(1565.7 \text{ ng m}^{-3})$ restaurants. The lowest concentration was found in the Szechwan Hotpot (299.6 ng m^{-3}). The concentration of BaP in Shaanxi Noodle was 247 ng m⁻³ on average, 2.5–11.6 times much higher than those in the other restaurants $(21.3-99.3 \text{ ng m}^{-3})$. BaP is often used as a marker for carcinogenic risk assessment. The BaP levels measured in all restaurants much exceeded the indoor guideline of 1.0 ng m^{-3} established by the WHO, suggesting an extremely high cancer risk to the dining area workers (i.e., waiter, waitress and cashiers) in restaurants. In term of Σ PAHs per capita, the highest value was also observed in Shaanxi Noodle, as the same as the findings for gaseous carbonyls. The contribution of coal combustion used as cooking fuel in its dining area should not be underestimated. Phenanthrene (PHE), chrysene (CHR), and benzo[a]anthracene (BaA) are the three most abundant PAHs among the restaurants, consistent with the cooking emission profiles reported by Rogge et al. (1991) and Schauer et al. (1999).

Diagnostic ratios of PAHs isomers have been used to identify potential pollution sources. The ratios of anthracene (ANT) to (ANT + PHE) > 0.10 and BaA / (BaA + CHR) > 0.35 suggest remarkable contributions from combustion sources to indoor dining areas (Tobiszewski and Namieśnik, 2012). The BaP to benzo[*ghi*]perylene (BghiP) was between 1.4–1.8 in the smoking restaurants, revealing a contribution of tobacco combustion (Katsoyiannis et al., 2007; Li et al., 2017). On average, the mass compositions of low molecular weight PAHs $(\Sigma PAHs_{\leq 178})$ (with 2–3 aromatic rings) (40.6%–65.7%) were higher than middle molecular weight PAHs ($\Sigma PAHs_{\geq 02-228}$) (with 4 aromatic rings) and high molecular weight PAHs ($\Sigma PAHs_{\geq 252}$) (with $\geq 5-6$ aromatic rings) in all sampled restaurants (Chen et al., 2012) (Fig. 2). Different from the thermal processes in coal-fired power plants and heating/industrial boilers, comparatively low cooking temperatures facilitate the formation of lighter PAHs (Zhang et al., 2016). In addition, the high contributions of low molecular weight PAHs, which mostly partitioned in gas-phase, could be attributed to their absorptions on the filter matrix during the sampling events. The largest distribution of high molecular weight PAHs was also seen in Shaanxi Noodle. Considering that the toxicity of a PAH ascends with an increase of molecular weight, the emissions from the coal combustion should be sufficiently contributed to health impact.

3.4. PM_{2.5}-bound heavy metals

Table 4 lists the concentrations of carcinogenic heavy metals in the four selected restaurants. Both Pb and Hg are neuro-developmental toxins at low exposure doses, and the inhalation of atmospheric particles is an important exposure pathway for them. The lead (Pb) concentrations ranged from 1720 to 2174 ng m⁻³, 5.6–7.1 higher than the annual average of 306 ng m⁻³ in ambient airs in Xi'an (Xu et al., 2012). Pb is the only metal listed in Chinese air quality standards due to its high toxicity to human. The annual maximum acceptable limits of Pb for total suspended particles (TSP, PM with aerodynamic diameters \leq 100 µm) outdoor and indoor is 1.0 µg m⁻³ set by the National Ambient Air Quality Standards (NAAQS) in China (GB 3095-2012). The Pb concentrations in the dining areas all exceeded the

Table 5

Cancer risk assessment results on Group 1 compounds.

Compounds	Szechwan hotpot	Hunan	Shaanxi noodle	Chinese barbecue	Chinese vegetarian	Korean barbecue	Italian	Indian
	$\times 10^{-4}$	$\times 10^{-4}$	$\times 10^{-4}$	×10 ⁻⁴	×10 ⁻⁴	$\times 10^{-4}$	$\times 10^{-4}$	$\times 10^{-4}$
Formaldehyde	0.27	0.50	0.35	1.46	0.44	0.75	0.26	0.66
Acetaldehyde	0.29	0.13	0.10	0.18	0.11	0.13	0.06	0.19
Cadmium	_ ^a	-	-	-	NA ^b	-	NA	0.23
Benzo[a]pyrene	0.03	0.08	0.37	0.05	NA	0.09	NA	0.15
Total	0.59	0.71	0.82	1.70	0.54	0.98	0.32	1.23

^a Values were below detection limit or insufficient filters for chemical analysis.

^b No data was collected due to sampler fault.

Concentrations of hydroxyl radical (ng m $^{-3}$) and PM (µg m $^{-3}$) in selected commercial restaurants.

Compounds	Szechwan hotpot	Hunan	Shaanxi noodle	Chinese barbecue	Korean barbecue	Indian
•OH (dining period)	149.6	116.3	145.6	294.4	163.7	248.5
•OH (non-dining period)	179.1	208.9	168.6	589.2	204.2	387.0
Non-dining/dining ratio	1.2	1.8	1.2	2.0	1.2	1.6
PM _{2.5} (dining period)	445.4	236.4	345.8	1057.9	429.7	590.0
PM _{2.5} (non-dining period)	617.2	319.2	881.7	1352.1	330.0	1051.7
Non-dining/dining ratio	1.4	1.4	2.5	1.3	0.8	1.8

guideline. Nickel (Ni) is another heavy metal widely concerned because of its carcinogenic potential. Its concentrations were 104-227 ng m⁻³ on average among the sampled restaurants. Our values are 17.6-38.4 times of the 10-year average (5.9 ng m⁻³) in the ambient airs in Xi'an (Xu et al., 2017). Currently, there is no any indoor standard established for Ni or other carcinogenic metals in China.

Fig. 3 compares the composition profiles for those carcinogenic metals quantified in this study. Pb is the most abundant element in all restaurants (>60%), followed by Hg (~20%), Cr, Ni, and Co. Cd was only detected in the samples collected in the Indian restaurant, while none of As and Se were detectable in all restaurants. Slightly difference on the absolute concentrations and mass per capita for each element or elemental compositions was obtained among the four restaurants. Ni is usually found in ETS, stainless-steel made kitchen utensils, and cookware (Kuligowski and Halperin, 1992). Higher Ni concentrations and compositions were thus found in the smoking dining units (i.e., Hunan and Chinese Barbecue restaurants). Those heavy metals (e.g., Pb) could be originated from the metallurgical cooking implements and tableware, which are operated at high temperatures (Siao and Rajasekhar, 2008). The anthropogenic inputs such as combustions of cooking fuels could also contribute to the emissions of the metallic elements such as Hg (Xu et al., 2012). Besides, the metallic pots and prods used in Hunan and Chinese Barbecue restaurants were more aged than those used in other two restaurants under the on-site inspections. They could be additional sources for the slightly higher levels of metals measured. However, no additional chemical tests (e.g., flame or emission tests and coating examinations) have been conducted on both cookware and fuels due to limitations in realistic environment.

3.5. Exposure cancer risk assessment

Cancer risk estimations for the pollutants are calculated and shown in Table 5. The slope factor of formaldehyde is 0.043 and 0.037 mg^{-1} kg⁻¹ d⁻¹ and of acetaldehyde is 0.0073 and 0.0063 mg⁻¹ kg⁻¹ d⁻¹, for male and female, respectively. The highest risk of formaldehyde (1.46×10^{-4}) was seen in Chinese Barbecue, while Italian restaurant had the lowest value of 0.26×10^{-4} . The cancer risks of acetaldehyde had the similar trend but were less significant compared with those of formaldehyde, on a range of 0.06×10^{-4} to 0.29×10^{-4} . For elemental, Cd is the single metal with the slope factor, with an estimated cancer risk of 0.23×10^{-4} in Indian restaurant. The highest cancer risk for BaP is found in Shaanxi Noodle (0.37×10^{-4}) , while the lowest is reported in Szechwan Hotpot (0.03×10^{-4}) . The results of cancer risk assessments demonstrate that the working environment of dining areas were hazard to health. Overall, Chinese Barbecue had the worst air quality while the Italian was the best among the sampled restaurants. Indian, Korean Barbecue and Hunan restaurants also reached the concern levels of 10⁻⁵ level (one in 100,000) for formaldehyde. It must be noted that Chinese barbecue is one of the popular dining in China. The poor air quality in its dining area should be a concern to both workers and diners.

3.6. Formation of hydroxyl radical (•OH) potential in lung fluid

Table 6 compares the concentration of PM_{2.5} mass and •OH formation concentrations in the six restaurants between dining and nondining periods. The PM collected in Chinese Barbecue generated has the highest concentration of •OH (294.4 ng m⁻³), following by Indian, Korean Barbecue, Szechwan Hotpot, Shaanxi Noodle and Hunan restaurants, which are in range of 116.3–248.5 ng m⁻³. The differences of the •OH concentrations could be attributed to the chemical compositions of PMs (Shen and Anastasio (2011)). Our results demonstrate that the cooking method is a controlling parameter for the generations. The barbecue types of restaurants could generate higher percentages of •OH from PM where grill is the dominated cooking method. The boilingdominated restaurants (i.e., Szechwan Hotpot and Shaanxi Noodle) had relatively low production of •OH, while the stir-fry restaurant had the lowest.

Different from other gaseous and particulate pollutants, the •OH formation concentration elevated in the non-dining section (i.e., after lunch time) (Table 6). Chinese Barbecue showed the largest raise on average which is double of that measured in the dining periods. Poor ventilation facilitates the accumulations of gaseous and PM pollutants indoor, leading of series of heterological reactions, particle aging, and formation of secondary pollutants. Secondary organic aerosol (SOA) is often as known as a mix of multiple-functional oxidized polar species that reasonably generate more •OH due to their high oxidizing potentials (Pope and Dockery, 2006). More experiments and studies are needed to further characterize and compare the chemical compositions of PM. However, the findings on •OH formation potential offer a new view on the health assessment rather than evaluation of the toxic substances levels.

4. Conclusions

Foods, ingredients, oils, cooking methods, fuels, implements and tableware can contribute to the variations of emission profiles for airborne carbonyls, particulate-bound PAHs and heavy metals. In-dining cooking activities and ETS can affect the indoor air quality of dining places as well. Even through the concentrations of formaldehyde and acetaldehyde in the dining areas were well below the guidelines, the particulate-bound BaP and Pb were much exceeded the acceptance levels recommended by the WHO. The indoor air quality and potential cancer risks of dining environments in commercial restaurants must be aware for both workers and diners. The elevation of •OH concentrations after the dining periods also act as an alert on the potential secondary formation of more reactive substances, which greatly impact on human's respiratory system under a low ventilation condition.

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