



## Chemical Characteristics of Fine Particles Emitted from Different Chinese Cooking Styles

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### ABSTRACT

Cooking process was regarded as one of the most significant contributor to fine particles (PM<sub>2.5</sub>) in ambient atmosphere and its chemical characteristics would be great different among various cooking styles. In this study, PM<sub>2.5</sub> emitted from four different Chinese cooking styles, including Home cooking, Shandong cuisine, Hunan cuisine, and Barbecue, were collected using a dilution sampling system. Then, PM<sub>2.5</sub> mass concentrations were weighted, and its chemical composition were analyzed. It was found that Barbecue emitted PM<sub>2.5</sub> concentrations with the highest level, followed by Home cooking, Shandong cuisine and Hunan cuisine. PM<sub>2.5</sub> emission amounts and emission factors were also estimated according to the measured data. Home cooking notably had the highest levels. The difference between Barbecue and other cuisines of PM<sub>2.5</sub> chemical profiles were the largest by using the coefficient of divergence (CD) method. The predominant chemical composition was organic carbon (OC) in PM<sub>2.5</sub>. The main water-soluble ions were Na<sup>+</sup>, SO<sub>4</sub><sup>2-</sup>, NO<sub>3</sub><sup>-</sup>, Cl<sup>-</sup>, and Ca<sup>2+</sup>, and Fe, S, and Ca had made up a big proportion of element. Little difference had been found between the mass fractions of n-alkanes and polycyclic aromatic hydrocarbons (PAHs) from different cooking cuisines. However, Barbecue displayed the highest mass fractions of organic acids in cooking fume.

**Keywords:** Cooking styles; PM<sub>2.5</sub>; n-Alkanes; PAHs; Organic acids.

### INTRODUCTION

Cooking fume was generated by edible oil and food after a series of complex chemical reaction such as the thermal oxidation and thermal cracking at a high temperature during the cooking process. It was regarded as one of the main sources of the urban atmospheric fine particles (PM<sub>2.5</sub>, particles with an aerodynamic diameter of  $\leq 2.5 \mu\text{m}$ ) in China (Kearney *et al.*, 2011; Wallace and Ott, 2011; Wheeler *et al.*, 2011), which contribution to Chinese national total fine particle mass was reported to be 7% by He *et al.* (2004). At present, cooking fume may be another significant source for PM<sub>2.5</sub> pollution besides industrial pollution and vehicle emissions in some cities (He *et al.*, 2004). Roe *et al.* (2005) also indicated that large scale cooking fumes might make a considerable contribution to PM<sub>2.5</sub> especially in urban environment for it generally contains a range of health-

damaging pollutants, such as heterocyclic amines and unsaturated aldehydes. Studies about adverse effects on human health from cooking fumes believed that women exposure to cooking-generated aerosol had the potential link with women's respiratory inflammation responses as women are primarily involved in cooking activities (Stabile *et al.*, 2015).

To the best of our knowledge, studies in the particle emissions from various anthropogenic sources have mainly focused on industrial pollution (Farao *et al.*, 2014; Institutu *et al.*, 2014; Wu *et al.*, 2014), and transportation source (Cheng *et al.*, 2013; Bozlaker *et al.*, 2014; Shen *et al.*, 2014). Studies in particles emissions from cooking are relatively few, which mainly focused on the particle size distribution and mass concentration of inorganic components (Hussein *et al.*, 2006; See and Balasubramanian, 2006). For instance, Wallace *et al.* (2008) conducted an intensive measurement and believed that the majority of the particles emitted from cooking were less than 10 nm with the peak concentration occurring around 5 nm to 6 nm. Yeung and To (2008) draw a conclusion that size distribution of particles followed a lognormal distribution and the diameter increased as cooking temperature increased. Attention was also focused on ultrafine particles (UFPs, diameter < 100 nm) from cooking as they can affect the human health more than larger particles. Buonanno *et al.* (2009) conducted an investigation to evaluate indoor

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UFPs emissions produced during grilling and frying, and it had been found that food, oil type, and temperature had a significant effect on cooking emission. The mass concentration of inorganic components was also reported. Kang *et al.* (2009) carried out a pilot study which indicated that elevated concentrations for toxic elements (Cd, As, and Pb) in the tents were found far higher than those in outdoor air in Beijing. However, chemical organic components, such as n-alkanes, polycyclic aromatic hydrocarbons (PAHs), and organic acids in  $PM_{2.5}$  emitted from cooking were rarely reported by researchers. And it was difficult to determine organic acids directly due to their polarity characteristics. It needed to make it convert into volatile esters through derivation methods, and then measured using chromatographic (Zheng *et al.*, 2000).

Previous studies have provided valuable information on the characteristics of particles emitted from cooking. However, the negative effects on environment and human health from cooking exacerbated with the fast growth of the quantities and scales of China's catering industry for nearly 20 years. Based on the information provided above, how to effectively control the cooking fume becomes increasingly urgent. In this study,  $PM_{2.5}$  emitted from different cooking styles, including Home cooking, Shandong cuisine, Hunan cuisine, and Barbecue, were collected by using a dilution sampling system and then analyzed its chemical composition, including 23 elements, 10 water-soluble ions, organic carbon (OC), elemental carbon (EC), 23 n-alkanes, 16 PAHs and 11 organic acids. Afterward,  $PM_{2.5}$  emission factors and emission amounts from cooking fume were estimated. Better understanding on the chemical characteristics of  $PM_{2.5}$  from cooking could provide the effective support

to improve air quality and ensure the residents health.

## METHODS

### Sample Collection

$PM_{2.5}$  emitted from stationary sources was generally categorized as primary fine particles and secondary fine particles (Robinson *et al.*, 2010). No standard method was released currently for  $PM_{2.5}$  collection from combustion sources in China (Li *et al.*, 2011). The traditional sampling method was designed for compounds that emitted directly from sources, and without secondary fine particles that cooled and diluted with ambient atmosphere. That may cause high uncertainty in  $PM_{2.5}$  estimate due to the high temperature of flue gas (Li *et al.*, 2011). The particle filter temperature should be at 42°C or lower according to dilution sampling standard proposed by International Organization for Standardization (ISO 25597:2013). The fume temperature was above 42°C at the vent of cooking exhausts measured in this study. Therefore, a dilution sampling technique was employed to collect  $PM_{2.5}$  from cooking fumes under real conditions (Fig. 1). The dilution sampling method utilizes a dilution chamber that mixes flue gas with dilution air to simulate the rapid cooling and dilution processes after hot flue gas exiting the exhaust pipe. Except primary fine particles, the condensation of particulate matter could also be collected simultaneously by using this method.

This system was suitable for field test, and mainly included the following modules: (1) Opium pipe was used for drainage the flue gas from cooking fume and real-time measurement of instantaneous flow rate and temperature from flue gas. (2) Heated inlet line was used to ensure the

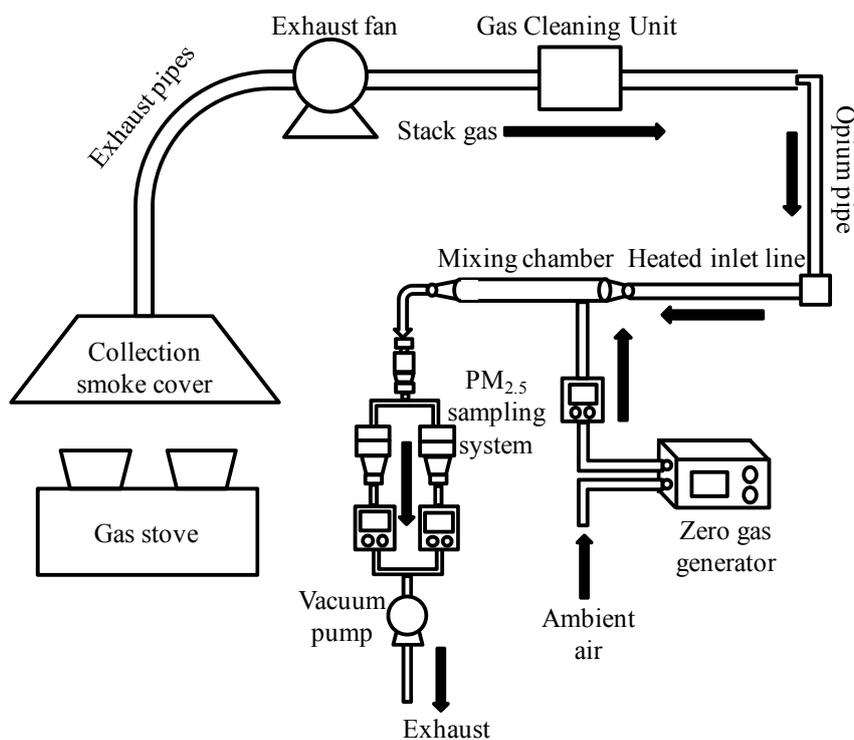


Fig. 1. Structure of dilution sampling system of cooking.

temperature of flue gas to prevent the temperature bring down during the gas from the opium pipe into the mixing chamber. (3) Zero gas generator was used to provide dilution air that had been filtered to remove particles. (4) In the mixing chamber, the flue gas was immediately diluted to create a drop in temperature to 42°C below, and the relative humidity was also below 70% RH. This could simulate the physical chemical process that may lead to the condensation of particulate matter. At the outlet of the mixing chamber, the samples were collected on the filters described later. During measurements, a micro SD was connected to the instrument, and the sampling time, flow rate, and dilution ratio could be exported after the sampling.

According to the Chinese emission standard of cooking fume (GB 18483-2001), the sampling point was located in the exhaust pipe on the vertical section on the rooftop of the building. During the measurements, oven was operated in the kitchen and extractor hood was continuously running. Dilution sampling system was used to collect PM<sub>2.5</sub> samples with a flow rate of 8.35 L min<sup>-1</sup>. Samples were collected simultaneously on 47-mm teflon and quartz filters (Whatman Inc Maidstone, UK), which were used for the analysis of elements/ions and OC/EC, respectively. The samples were collected during two peak cooking periods, 10:30-13:00 and 17:00–20:00. Each restaurant was measured in three consecutive days, and a total of 25 measurements were conducted. Filters were all weighed using a One Over Ten-thousand Analytical Balance (Sartorius TB-215D, Germany) before and after sampling under constant temperature (20 ± 5°C) and relative humidity (40 ± 2% RH) (Cheng *et al.*, 2013). Filters were placed in zip lock bags immediately after sample collection, and stored in a refrigerator at about 4°C until chemical analysis to avoid the evaporation of volatile components. The ambient PM<sub>2.5</sub> mass concentrations were obtained from automatic air monitoring system, which could reflect the environmental quality of urban area.

This study selected four types of Chinese cooking cuisines which were quite popular in China, i.e., Home cooking, Shandong cuisine, Hunan cuisine, and Barbecue. Five commercial restaurants were selected in this study, two for Shandong cuisine and one for other cuisines. In details, three cuisines serve 360 days per year and have the cooking operation for 6 hours per day approximately. Barbecue is open 200 days per year and 7 hours per day. The basic information of each cooking styles was summarized in Table 1, such as attendance, features, fuel, oil types, and gas cleaning unit. Various gas cleaning units had been made to reduce emissions from cooking in China, including media filters, carbon filters and electrostatic precipitators. In this study, the filters installed in Home cooking, Shandong cuisine, and Hunan cuisine was the same type—static-type lampblack purifier. And the removal rate of cooking fumes was above 95%, which follows up with the requirements of Chinese emission standard of cooking fume (GB 18483-2001). However, Barbecue was not installed purification device.

#### **Chemical Analysis of Elements**

One-fourth of teflon filter was digested at 170°C for 4 h in

**Table 1.** Parameters of sampling restaurants.

Cooking styles	Attendance	Features	Fuel	Oil types	Gas cleaning unit
Home cooking	80%–90%	Stir-frying, stewing and steaming	Natural gas	Salad oil	Static-type lampblack purifier
Shandong cuisine	80%–90%	Fresh and light taste	Natural gas	Soybean oil	Static-type lampblack purifier
Hunan cuisine	70%–80%	Spicy, sour taste	Liquefied gas	Salad oil	Static-type lampblack purifier
Barbecue	80%–90%	Roast, deep-frying, baked, fried, the main ingredients for cattle and sheep	Fruit charcoal	Salad oil	None

high-pressure teflon digestion vessel with 3 mL concentrated HNO<sub>3</sub>, 1 mL concentrated HClO<sub>4</sub>, and 1 mL concentrated HF (Han *et al.*, 2007). After cooling, the solutions were dried, and then diluted to 10 mL with distilled-deionized water. Finally, the concentrations of 23 elements (Mg, Al, S, Ca, Sc, Ti, V, Cr, Mn, Fe, Co, Ni, Cu, Zn, As, Se, Sr, Mo, Cd, Sb, Ce, Eu, and Pb) were determined by Inductively Coupled Plasma Mass Spectrometry (ICP-MS, 7500a, Thermo).

#### **Chemical Analysis of Water-Soluble Ions**

One-fourth of teflon filter was extracted ultrasonically by 10 mL distilled-deionized water and oscillated for 40 min in supersonic cleaner (Wang *et al.*, 2012). The concentrations of 10 water-soluble ions (F<sup>-</sup>, Cl<sup>-</sup>, NO<sub>3</sub><sup>-</sup>, SO<sub>4</sub><sup>2-</sup>, NO<sub>2</sub><sup>-</sup>, Na<sup>+</sup>, NH<sub>4</sub><sup>+</sup>, K<sup>+</sup>, Ca<sup>2+</sup>, and Mg<sup>2+</sup>) were determined by Ion Chromatograph (Metrohm861 Advanced Compact IC, Switzerland). The detailed water-soluble ions analytical procedures were given elsewhere (Wang *et al.*, 2005).

#### **Chemical Analysis of OC and EC**

Carbonaceous species (OC and EC) were determined by the thermal/optical carbon analyzer (DRI Model 2001A, Desert Research Institute of United States). Firstly, OC was volatilized from the samples as the temperature gradually rose to 120°C (OC1), 250°C (OC2), 450°C (OC3), and 550°C (OC4) in a 100% helium atmosphere. Then the analyzer oven temperature was gradually ramped to 550°C (EC1), 700°C (EC2), and 800°C (EC3). The optical pyrolyzed carbon (OP) is also formed in this process. Carbon in these volatile components was converted into CO<sub>2</sub> by oxidant (MnO<sub>2</sub>), then converted into CH<sub>4</sub> through a converter, and was finally quantified with a flame ionization detector (FID). OC was defined as OC1 + OC2 + OC3 + OC4 + OP and EC as EC1 + EC2 + EC3 – OP according to the IMPROVE protocol (Chow *et al.*, 2004). Replicate analysis was conducted using blank filters, and the average value was used as the background concentration (Feng *et al.*, 2009).

#### **Chemical Analysis of Organic Matter Samples Extraction**

The organic matter determined in this study included 16 PAHs, 23 n-alkanes, and 11 organic acids. The quartz filters were cut into pieces and extracted 4 times with 50 mL of 1:1 v/v dichloromethane/acetone each for 20 min. The combined extracts were concentrated to 1–2 mL by rotary evaporation under gentle vacuum, and further reduced to 50 µL under a high-purity nitrogen gas stream (Zheng *et al.*, 2014). These concentrated filtrates were divided into three fractions, and spiked with hexamethylbenzene (99%, Supelco, USA) for the internal standards. The fraction for measuring acids was also derivatized with 14% BF<sub>3</sub>/CH<sub>3</sub>OH to convert organic acids to the corresponding methyl esters.

#### **GC-MS Analysis**

Three types of target compounds were analyzed using a gas chromatography - mass spectrometry (GC - MS, Model HP 5890II/5972, Dikma) equipped with a DB-5MS capillary column (30 m × 0.25 mm × 0.25 µm). The temperature program for the column oven was as follows: PAHs: the

initial temperature was hold at 80°C for 2 min, increased to 290°C at a rate of 5 °C min<sup>-1</sup> for 8 min, and then to 310°C at 5 °C min<sup>-1</sup> for 2 min. Alkanes: the initial temperature was hold at 50°C for 5 min, increased to 315°C at a rate of 12 °C min<sup>-1</sup> for 20 min. Organic acid: the initial temperature was hold at 50°C for 5 min, increased to 315°C at a rate of 5 °C min<sup>-1</sup> for 10 min. The sample was injected 1 µL in splitless mode, and high-purity helium (99.999%) with flow rate of 1.0 L min<sup>-1</sup> was used as the carrier gas. PAHs and alkanes were identified and quantified based on the retention time and selected ions mode (SIM), while organic acid was full scan mode from 30 to 700 amu at 4.00 scans per second.

The quantification of compounds was accomplished by using the following standards: (1) the mixed standard sample of precedent-controlled 16 PAHs (Chem Service, USA) (Zhang *et al.*, 2006); (2) n-alkanes (Chem Service, USA); (3) organic acids (Chem Service, USA). Then, the 7-point calibration curve method and internal standard method were adopted to quantify the target compounds. The regression coefficient for each component was all higher than 0.99, which showed the good linearity between the integral areas of peaks and the corresponding concentrations.

#### **Quality Control**

In order to evaluate the possible contamination and loss during the pretreatment including ultrasonic extraction, separation and purification, filter blanks, solvent blanks and the standard recovery indicators were analyzed to check the background under the same experimental procedures. The concentrations of detected compounds in the filter blanks and solvent blanks were below the detection limits. Therefore, the results were not corrected for field blanks. The recoveries of each target compound were in the range of 80% to 120% during extraction and analysis. The relative standard deviation of standard compounds was within 10%.

## **RESULTS AND DISCUSSION**

#### **PM<sub>2.5</sub> Mass Concentrations Emitted from Different Cooking Cuisines**

Table 2 demonstrated the PM<sub>2.5</sub> mass concentrations in the cooking fume and ambient air. It was found that the PM<sub>2.5</sub> emitted from cooking fume were 494.9 ± 240.6 µg m<sup>-3</sup>, 404.8 ± 192.3 µg m<sup>-3</sup>, 331.3 ± 179.0 µg m<sup>-3</sup>, and 1841.9 ± 763.7 µg m<sup>-3</sup>, respectively for Home cooking, Shandong cuisine, Hunan cuisine and Barbecue. The PM<sub>2.5</sub> concentrations in cooking fumes were 4.3 to 20.2 times higher than their ambient concentrations of the corresponding sampling time, which implied a greater influence on the atmospheric PM<sub>2.5</sub>.

The cooking method has been identified as one of the important factors affecting emission of some contaminants (Abdullahi *et al.*, 2013). PM<sub>2.5</sub> concentration from cooking Hunan cuisine dominated by stir-frying and blasting was believed higher than Home cooking due to the difference of cooking style by See and Balasubramanian (2006). The opposite conclusion was found in this study. That mainly due to that the attendance of Hunan cuisine was lower, and the status of smoke purifier was good, which was relatively new equipment and dust removal effect was better. It was

**Table 2.** Comparison of the average PM<sub>2.5</sub> mass concentration between cooking emissions and ambient air (μg m<sup>-3</sup>).

Cooking styles	PM <sub>2.5</sub> concentration of cooking emissions	PM <sub>2.5</sub> concentration of ambient air	Ratio
Home cooking	494.9 ± 240.6	115.5	4.3
Shandong cuisine	404.8 ± 192.3	58.5	6.9
Hunan cuisine	331.3 ± 179.0	48.4	6.8
Barbecue	1841.9 ± 763.7	91.4	20.2

also found that Barbecue had the highest PM<sub>2.5</sub> concentration, and 3.7 to 5.6 times higher than other cooking styles. The observation was consistent with McDonald *et al.* (2003) that charbroiling produced higher concentrations than frying. This may be generally attribute to that Barbecue was fueled by fruit charcoal which might produce more fine particles in the combustion process. Additionally, the fume from Barbecue emissions was discharged into the environment atmosphere directly without particulate control devices compared to other cooking styles, so more fine particles were produced. Therefore, purification measures with high removal efficiency might be the effective measure to alleviate the PM<sub>2.5</sub> pollution released from cooking fumes.

PM<sub>2.5</sub> emission amounts (EA) and emission factors (EF) for cooking process were further calculated by Eqs. (1) and (2).

$$Q = 10^{-9} \times C_{\text{PM}_{2.5}} \times S \times v \times t \quad (1)$$

$$EF = \frac{10^8 \times Q}{u} \quad (2)$$

where Q represents the annual PM<sub>2.5</sub> EA of a certain restaurant (kg a<sup>-1</sup>), C<sub>PM<sub>2.5</sub></sub> were the measured mass concentration of PM<sub>2.5</sub> (μg m<sup>-3</sup>) emitted from cooking fume. S, v, and t represent stack area (m<sup>2</sup>), stack outlet velocity (m s<sup>-1</sup>) and cooking operation time per year (h a<sup>-1</sup>), respectively. U represents the amount of cooking oil consumed annually by the restaurant (kg a<sup>-1</sup>). And EF was the PM<sub>2.5</sub> EF based on the oil consumption (g PM<sub>2.5</sub>/kg oil consumed).

We investigated the information of stacks, and annual cooking operation time for each restaurant, and then calculated their annual PM<sub>2.5</sub> EA and EF based on the PM<sub>2.5</sub> concentrations in the fume measured. Table 3 gave the summary. The number of customers served for each sampling was also recorded to calculate the EF. The amount of cooking oil used by each person was assumed 8.84 kg one year (EPA, 2013). For the PM<sub>2.5</sub> EA, Home cooking studied in this paper notably had the highest emission (12.4 kg a<sup>-1</sup>) than other cooking styles, especially for Barbecue which had the lowest emission (3.8 kg a<sup>-1</sup>). It could be found that the emission of Shandong cuisine was basically consistent with Hunan cuisine, which was 1.6–1.8 times higher than that of Barbecue. Even though the PM<sub>2.5</sub> concentration of Barbecue was high, the lower cooking time and stack area leading to the low PM<sub>2.5</sub> EA. For EF, the distribution of Home cooking had been observed in the same order as PM<sub>2.5</sub> EA, which had the highest EF (0.039 g kg<sup>-1</sup>). The other three kinds of cuisines showed a lesser margin (0.019 g kg<sup>-1</sup>–0.027 g kg<sup>-1</sup>).

### Chemical Composition of PM<sub>2.5</sub> Emitted from Different Cooking Cuisines

The chemical profiles of PM<sub>2.5</sub> emitted from different cooking cuisines were concluded in Table 4. As expected, OC constituted the largest fraction of PM<sub>2.5</sub> from cooking, accounting for 36.2%–42.9% of the fine particle mass, which was consistent with data reported by Abdullahi *et al.* (2013) and See and Balasubramanian (2008). In contrast, the EC accounted for 0.8%–1.3% of the PM<sub>2.5</sub> mass concentration except Barbecue. Several water soluble ions were also measured in the fine particle emissions at a lower but noticeable percentages, which made up of about 9.1%–17.5% of the total PM<sub>2.5</sub> mass concentration. The main ions components were Na<sup>+</sup>, SO<sub>4</sub><sup>2-</sup>, K<sup>+</sup>, Cl<sup>-</sup>, and Ca<sup>2+</sup>, which accounting for 62.9%–85.7% of total water soluble ions. According to previous research by the Public Utilities Board (PUB) in Singapore, the quantity of SO<sub>4</sub><sup>2-</sup> and F<sup>-</sup> may generally attributable to the presence in potable water (PUB, 2007). NO<sub>3</sub><sup>-</sup> was believed to be derived from NO<sub>x</sub> emissions from the gas stoves (Arbex *et al.*, 2007), and the abundance of Na<sup>+</sup> and Cl<sup>-</sup> was probably derived from salt. Barbecue with higher concentration of K<sup>+</sup> might be from roast emissions (Watson and Chow, 2001). For another, inorganic elements was found to contribute about 7.3%–12.0% of the total PM<sub>2.5</sub> mass concentration due to their greater presence in cooking oil and raw materials (Anwar *et al.*, 2004). Fe, S, and Ca had made up a big proportion in PM<sub>2.5</sub>. Fe, Cr, and Ni might from top-of-the-stove stainless steel cooking ware under a high temperature during cooking, where contained a certain amount of chromium alloy elements (Kumar *et al.*, 1994). However, other elements contented in PM<sub>2.5</sub> were scarce.

The major chemical feature in the four cooking cuisines was similar, but the content of species among different cuisines might be difference due to various cooking styles. In order to compare the differences in chemical profiles of PM<sub>2.5</sub> quantitatively, coefficient of divergence (CD) method was used to analyze the level of similarity of PM<sub>2.5</sub> emitted from different cuisines. The CD can be calculated as follows (Ji, 2006):

$$CD_{jk} = \sqrt{\frac{1}{p} \sum_{i=1}^p \left( \frac{x_{ij} - x_{ik}}{x_{ij} + x_{ik}} \right)^2} \quad (3)$$

where j and k were the different cuisines, CD<sub>jk</sub> represents the CD between cuisine j and cuisine k, X<sub>ij</sub> and X<sub>ik</sub> represents the concentration of a certain PM<sub>2.5</sub> species i in cuisine j and k (μg m<sup>-3</sup>), p represent the number of chemical species in the calculation.

**Table 3.** PM<sub>2.5</sub> emission amounts and emission factors of four cooking cuisines.

Parameters	Home cooking	Shandong cuisine	Hunan cuisine	Barbecue
PM <sub>2.5</sub> concentration (μg m <sup>-3</sup> )	494.9	404.8	331.3	1841.9
Stack outlet velocity (m s <sup>-1</sup> )	9.0	5.3	7.5	4.5
Stack area (m <sup>2</sup> )	0.36	0.36	0.36	0.09
Cooking operation time (h a <sup>-1</sup> )	2160	2160	2160	1400
PM <sub>2.5</sub> EA (kg a <sup>-1</sup> )	12.4	6.0	6.9	3.8
PM <sub>2.5</sub> EF (g kg <sup>-1</sup> )	0.039	0.019	0.027	0.021

**Table 4.** Chemical profiles of PM<sub>2.5</sub> emitted from different cooking styles (%).

Species	Home cooking		Shandong cuisine		Hunan cuisine		Barbecue	
	Average ± SD		Average ± SD		Average ± SD		Average ± SD	
OC	40.7	6.6	42.8	17.9	42.9	9.4	36.2	6.6
EC	1.3	0.6	0.9	0.3	0.8	0.4	18.4	12.0
F <sup>-</sup>	0.1	0.0	0.1	0.1	0.3	0.3	0.1	0.2
Cl <sup>-</sup>	1.0	0.5	1.0	0.5	2.1	2.2	2.2	2.1
NO <sub>2</sub> <sup>-</sup>	0.0	0.1	0.0	0.1	0.1	0.1	0.0	0.0
NO <sub>3</sub> <sup>-</sup>	0.1	0.1	0.2	0.2	1.2	1.2	0.3	0.3
SO <sub>4</sub> <sup>2-</sup>	2.3	1.2	2.4	3.1	2.7	1.5	1.5	1.2
Na <sup>+</sup>	1.8	1.1	1.9	1.2	3.0	0.1	1.4	1.0
NH <sub>4</sub> <sup>+</sup>	1.9	1.5	0.9	0.9	2.6	2.7	0.5	0.7
K <sup>+</sup>	1.9	0.8	2.3	2.9	1.8	0.8	2.4	1.0
Ca <sup>2+</sup>	0.7	0.4	2.3	1.4	1.4	0.6	0.3	0.4
Mg <sup>2+</sup>	1.8	1.0	2.4	1.7	2.3	1.8	0.4	0.5
Mg	0.3	0.2	0.4	0.3	0.3	0.2	0.6	0.6
Al	0.5	0.2	0.6	0.4	0.5	0.2	0.5	0.3
S	4.1	1.4	4.6	2.7	2.4	1.5	3.0	2.7
Ca	1.3	1.2	1.9	1.4	1.0	0.6	1.6	1.8
Ti	0.2	0.2	0.2	0.1	0.2	0.1	0.2	0.2
Cr	0.3	0.2	0.5	0.3	0.3	0.1	0.2	0.3
Mn	0.0	0.0	0.1	0.1	0.0	0.0	0.0	0.0
Fe	3.2	3.2	3.2	2.8	2.5	1.3	1.0	0.5
Ni	0.3	0.2	0.2	0.2	0.2	0.1	0.1	0.1
Cu	0.0	0.0	0.1	0.1	0.0	0.0	0.0	0.0
Zn	0.1	0.1	0.2	0.3	0.1	0.0	0.1	0.1

Notes: The content of Sc and other inorganic elements were not detected.

Theoretically, CD ranges from 0 to 1. The CD value of 0 means the same chemical profiles between two cuisines, while when CD arrives at 1, it means that the chemical profiles of two cuisines were entirely different. This study obtained CD = 0.3 as demarcation point of similarity of PM<sub>2.5</sub> chemical profiles or not according to the result studied by Ji (2006). Table 5 demonstrated the averaged CD of PM<sub>2.5</sub> chemical profiles between different cooking cuisines, which ranged from 0.25 to 0.44. The difference between Barbecue and other cuisines were the largest with CD exceeding 0.35. However, the obvious difference among Home cooking, Shandong cuisine, and Hunan cuisine was as never found.

#### Particulate Organic Compounds from Different Cooking Cuisines

As discussed above, OC accounted for a larger proportion of PM<sub>2.5</sub> concentration emitted from cooking fume. Therefore, organic components were further analyzed, mainly including n-alkanes, PAHs and organic acids. The mass fractions of organic compounds in PM<sub>2.5</sub> from different cooking cuisines

(ng μg<sup>-1</sup>) were demonstrated in Table 6.

Little difference had been found between the mass fractions of n-alkanes in PM<sub>2.5</sub> from four cooking cuisines studied in this paper (581.4 ng μg<sup>-1</sup>–661.2 ng μg<sup>-1</sup>). In addition, the higher mass fractions of n-alkanes occurred at C16–C26, which accounted for 61.8%–69.0% of the total n-alkanes.

The formation of PAHs was mainly derived from the nature organic matter of cooking oil during incomplete combustion (Zhao *et al.*, 2007). It could be found in Table 6 that the 16 PAHs comprised only a minor fraction (40.4 ng μg<sup>-1</sup>–47.7 ng μg<sup>-1</sup>) of the total mass PM<sub>2.5</sub>. The similar distribution of PAHs components had been observed from different cooking cuisines, and the dominant components were naphthalene, phenanthrene, fluoranthene, pyrene and fluorene, constituting 77.1%–84.1% of the total quantified PAHs compounds. According to the 2nd-level air quality in China (GB 3095-2012), the average daily benzo[a]pyrene concentration should be less than 2.5 ng m<sup>-3</sup>. However, benzo[a]pyrene emitted from Barbecue was 8.8 times higher than this limitation value during the sampling period,

**Table 5.** CD of PM<sub>2.5</sub> chemical profiles from different cuisines.

CD	Home cooking	Shandong cuisine	Hunan cuisine	Barbecue
Home cooking	0			
Shandong cuisine	0.25	0		
Hunan cuisine	0.28	0.29	0	
Barbecue	0.35	0.44	0.42	0

**Table 6.** Mass fractions of organic compounds in PM<sub>2.5</sub> from different cooking cuisines (ng μg<sup>-1</sup>).

	Species	Home cooking	Shandong cuisine	Hunan cuisine	Barbecue
n-alkanes	C8	22.5	7.0	11.4	12.3
	C9	3.4	2.9	3.7	2.2
	C10	13.1	11.4	14.3	14.7
	C11	7.7	7.4	10.6	13.2
	C12	14.7	16.2	13.0	21.4
	C13	2.3	2.6	4.1	3.8
	C14	18.4	22.8	21.4	27.6
	C15	12.6	14.7	22.1	15.8
	C16	36.3	44.4	61.1	44.5
	C17	57.5	61.9	90.4	58.3
	C18	52.7	55.4	67.5	44.5
	C19	36.8	34.3	35.3	30.6
	C20	51.1	48.0	45.6	45.7
	C22	66.9	61.5	62.6	61.2
	C24	47.8	49.9	45.2	45.2
	C26	53.7	45.8	45.4	48.8
	C28	35.3	31.0	32.2	35.0
	C30	25.1	22.5	23.7	27.4
	C32	16.4	15.8	15.5	19.9
	C34	10.2	9.8	9.6	13.7
C36	7.2	6.9	7.0	9.9	
C38	4.7	4.6	10.6	8.9	
C40	3.7	4.6	8.9	8.7	
	<b>Total mass</b>	<b>600.1</b>	<b>581.6</b>	<b>661.1</b>	<b>613.2</b>
PAHs	Naphthalene	21.8	19.4	16.1	27.1
	Acenaphthylene	0.4	0.3	0.7	0.5
	Acenaphthene	0.5	0.5	0.7	0.6
	Fluorene	1.8	1.5	2.1	1.6
	Phenanthrene	7.5	6.4	7.8	4.8
	Anthracene	0.3	0.3	0.6	0.3
	Fluoranthene	5.2	4.4	4.5	2.7
	Pyrene	2.3	2.2	2.0	1.3
	Benz[a]anthracene	1.0	1.0	1.2	1.4
	Chrysene	1.2	1.2	1.2	1.1
	Benzo[b]fluoranthene	1.1	0.9	1.4	2.1
	Benzo[k]fluoranthene	0.6	0.4	0.8	0.6
	Benzo[a]pyrene	0.4	0.3	0.7	1.2
	Indeno[1,2,3,c,d]pyrene	0.7	0.6	0.9	1.0
	Benzo[g,h,i]perylene	1.1	0.7	1.1	1.0
Dibenz[a,h]anthracene	0.3	0.3	0.6	0.4	
	<b>Total mass</b>	<b>46.2</b>	<b>40.4</b>	<b>42.1</b>	<b>47.6</b>
Organic acids	Valtric acid	nd	nd	nd	nd
	Caproic acid	296.7	325.3	324.9	176.5
	Heptanoic acid	nd	nd	nd	nd
	Caprylic acid	376.2	312.3	398.0	476.0
	Pelargonic acid	295.5	441.7	569.4	249.4
	Capric acid	nd	nd	nd	nd

Notes: nd: not detected.

**Table 6.** (continued).

	Species	Home cooking	Shandong cuisine	Hunan cuisine	Barbecue
Organic acids	n-Hendecanoic acid	nd	nd	nd	nd
	Lauric acid	125.5	127.4	196.2	129.9
	Myristic acid	216.8	212.1	333.7	480.1
	Palmitic acid	1478.3	1281.2	2083.1	7982.9
	Stearic acid	1252.4	1030.1	1799.5	6861.0
	<b>Total mass</b>	<b>4041.3</b>	<b>3730.2</b>	<b>5704.8</b>	<b>16355.7</b>

Notes: nd: not detected.

which might had a great contribution on the carcinogens in atmospheric environment. In addition, benzo[a]pyrene emitted from Home cooking and Hunan cuisine also approach the limitation value, which was  $2.1 \text{ ng m}^{-3}$  and  $2.4 \text{ ng m}^{-3}$ , respectively. It was also found that Barbecue with deep-frying cooking style generated the highest concentration of PAHs, which were 3.8–6.3 times higher than other cooking styles. This observation was consistent with that reported by See and Balasubramanian (2008).

The highest component of organic compounds in the four cooking cuisines was organic acids compared to n-alkanes and PAHs, accounted for 85.7%–96.1% of the quantified organic compounds. That might due to acids were easily produced by thermal oxidation during the cooking process. It could be also found in Table 6 that organic acids emitted from Barbecue displayed the highest mass fractions in cooking fume, followed by Hunan cuisine, Home cooking, and Shandong cuisine. This probably suggests that different cooking method and raw materials affect the chemical composition of organic acids. Palmitic acid and stearic acid were the two most abundant organic acids in cooking fumes, accounting for 62.0%–90.8% of the total organic acids detected in this study. In addition, palmitic acid and stearic acid from Barbecue were 3.8–5.4 and 3.8–6.7 times higher than other cuisines, respectively. It could be due to that those two species were considered as possible tracers of meat cooking (Rogge *et al.*, 1991). That impacted significantly by the cooking ingredients of Barbecue that the main ingredients for beef and mutton contain large amounts of saturated acid (He *et al.*, 2004).

## CONCLUSIONS

The  $\text{PM}_{2.5}$  and its chemical composition emitted from different cooking cuisines, namely Home cooking, Shandong cuisine, Hunan cuisine, and Barbecue were collected using a dilution sampling system. The results showed that Barbecue gave the highest  $\text{PM}_{2.5}$  mass concentrations of cooking fume.  $\text{PM}_{2.5}$  concentrations from cooking fume were 4.3 to 20.2 times higher than ambient air. According to measured data,  $\text{PM}_{2.5}$  emission amounts (EA) and emission factors (EF) were estimated. Home cooking had the highest EA, followed by Hunan cuisine, Shandong cuisine, and Barbecue. As for EF, Home cooking also displayed the highest value, and the other three cuisines showed a lesser margin. OC was the major constituent and accounted for 36.2%–42.9%, while the concentration of EC was low. The water soluble ions had made up of about 9.1%–17.5% of the  $\text{PM}_{2.5}$ , and

$\text{Na}^+$ ,  $\text{SO}_4^{2-}$ ,  $\text{K}^+$ ,  $\text{Cl}^-$ , and  $\text{Ca}^{2+}$  occurred in greater quantities. Fe, S, and Ca had made up a big proportion in inorganic elements, while other elements contented were scarce. The coefficient of divergence (CD) method was applied to analyze the level of similarity of  $\text{PM}_{2.5}$ . The difference between Home cooking, Shandong cuisine, and Hunan cuisine were small except Barbecue. Little difference had been found between the mass fractions of n-alkanes and PAHs from different cooking cuisines. The higher mass fractions of n-alkanes occurred at C16–C26, and the dominant components of PAHs were naphthalene, phenanthrene, fluoranthene, pyrene and fluorine. Compared to n-alkanes and PAHs, organic acids accounted for a significant proportion of organic compounds, and Barbecue displayed the highest mass fractions in cooking fume. Palmitic acid and stearic acid were the two most abundant organic acids, accounting for 62.0%–90.8% of the total organic acids.

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