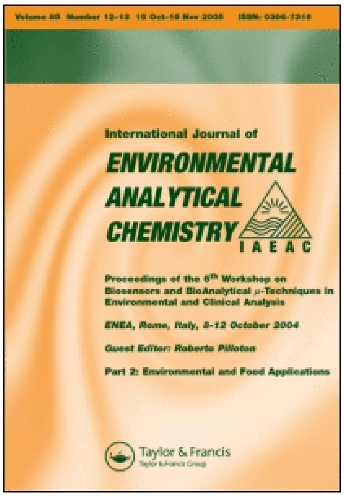

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The emission characteristics of hydrocarbon from Chinese cooking under smoke control

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The emission characteristics of hydrocarbon from Chinese cooking under smoke control

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To understand the hydrocarbon emissions in China, the source profile and emission factors of hydrocarbon species from cooking sources were measured. The measured commercial restaurants represented Sichuan cuisine, Zhejiang cuisine, household-style cuisine and Korean-style barbecue, respectively. Forty-one hydrocarbon species were quantified, in which the abundant species were n-butane, iso-butane, 2-methlybutane, pentane, propene, 1-butene, toluene and benzene. The hydrocarbon mixing ratios from Korean barbecue were evidently higher than those from the other Chinese-style restaurants, and the Sichuan-style restaurants had significantly higher emissions of iso-butane and d-limonene. The hydrocarbon source profiles of Chinese cooking were different from the profiles obtained from cooking sources of other cities. While the Korean-style barbecue had abundant species such as light alkanes, propene, benzene and toluene, similar to results for Western barbecue source, the relative percentages of these species were different, probably due to the difference in fuels for cooking. The hydrocarbon emission factors were estimated for Sichuan-style cuisine, which was 0.21g hydrocarbon per kg of meat cooked, much lower than that from the barbecue source.

Keywords: hydrocarbon speciation; source profile; emission factor; cooking

1. Introduction

As the precursor for the production of secondary organic aerosols (SOA) and also one of the precursors for the formation of ground-level ozone, hydrocarbons play very important roles in the atmospheric chemical processes. The accurate understanding of hydrocarbon sources will be relevant to formulate efficient secondary pollution abatement measures. Therefore, much effort has been placed on hydrocarbon source inventories [1–4] as well as source apportionment by receptor models [5–7]. For both approaches, the measurements of source profiles and emission factors of local hydrocarbon sources are essentially important to reduce the uncertainties in quantifying source emissions. A sizeable number of studies have been performed to investigate the hydrocarbon speciation emitted from sources such as vehicular exhaust, solvent evaporation, petrochemical industries, and

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biomass burning, etc. [5,6,8–10], but the measurements on hydrocarbon emissions from cooking sources are very rare.

The cooking activities could be an important source for hydrocarbon emission due to the very rapid increase of commercial restaurants and household kitchens in China. For example, Beijing city had 73,232 commercial restaurants in 2003, and rapidly increased to more than 150,000 in 2008 (http://www.ocn.com.cn/reports). Apart from these legal ones, there are a large number of on-street barbecues and hotpots that are not accounted into commercial sector. The hydrocarbon emissions from chain-driven charbroilers have been under variable control in some regions of the United States such as the South Coast Air Quality Management District (SCAQMD) [11–13]. China sets limits only for the concentrations of smoke aerosols from cooking activities, and hydrocarbon emissions are not controlled [14]. In fact, the limits on aerosols are not very efficiently implemented, and the hydrocarbon control devices are not required in the cooking smoke treatment facilities. What is more, hydrocarbon in oil smoke from on-street floating cooking units is emitted without any control.

The current studies on hydrocarbon source profiles and emission factors are mainly from United States, focusing on commercial cooking of meat [12]. A study aimed at establishing the method for emission test and emission factors of particulate matter (PM) and reactive organic gases (ROG) from commercial cooking operations has been performed since 1994 in the South Coast Air Quality Management District (SCQMD) by the Center for Environmental Research and Technology, Riverside Bourns College of Engineering, the University of California [15]. In the Northern Front Air Quality Study, the gaseous organic compounds source profile of meat cooking was established and used in a receptor model [16]. Emissions from street vendors have been studied in United States specifically for charcoal barbecues with different types of meat [17]. Schauer conducted a series of studies on emissions from hamburger cooking, food-frying operations and meat charbroiling [18–20]. The hydrocarbon source profiles from four main types of cooking in Mexico City were measured by Mugica *et al.* [21]. The impact of cooking operations on indoor air quality is also an issue that has been quite intensively investigated [22–24]. Because China has such a big diversity in cooking styles, and the raw material, oils and fuels used for cooking might be different from other countries, the hydrocarbon source profiles could be also different. The first work on the hydrocarbon source profiles measured in China was available recently [10], but unfortunately the source profiles of cooking operations were not included in the work.

This work is designed to perform the measurements on hydrocarbon speciation from typical commercial cooking operations in Beijing city, and to set up hydrocarbon source profiles for cooking sources. The results will be useful for hydrocarbon source apportionment researches in China. This work also measures hydrocarbon emission factors for one type of Chinese cooking. These could provide necessary data for hydrocarbon emission control from cooking activities in China.

2. Experimental

2.1 *The selection of restaurants*

Samples were taken at four large or middle-sized commercial restaurants in Beijing from 18 July to 30 July 2005. These restaurants were a Korean-style meat charbroiling restaurant and three Chinese cooking restaurants, including a Sichuan-style one, a Zhejiang-style one and a household-style one. The detailed cooking conditions are listed in Table 1. The Sichuan-style restaurants are the most popular ones, accounting for about 30% of the commercial restaurants in Beijing city. While Zhejiang-style restaurants represent the relative 'slap-up' ones including Shanghai-style and Guangzhou-style, they account for about 10% of the commercial restaurants. The Korean restaurants have become more and more popular in Beijing and account for about 5% of the commercial restaurants.

All the four restaurants had a ventilation system with cooking smoke treatment facilities, and the Korean restaurant was equipped with a hood over each charbroiling table and the oil smoke was drawn to a centralised pipe. See Table 2 for a specific description of emission control facilities.

Table 1. Comparison of the four restaurants selected for hydrocarbon emission measurement.

No.	Cooking style	Fuel	Cooking style description
1	Sichuan	Natural gas[a]	Heavy and lengthy cooking, meals are greasy and spicy
2	Zhejiang	Natural gas[a]	Light cooking, meals are fresh and salubrious
3	Household	Natural gas[a]	Similar to Sichuan style cooking
4	Korean charbroiling	Charcoal and natural gas[a]	Mainly charbroiling and frying of pork and beef
		Ingredient	Cooking oil (consumption, kg per day)
1	Sichuan	Meat and fish	Rapeseed oil, salad oil mix[b] (46 totally)
2	Zhejiang	Vegetable and seafood	Soy bean oil (90)
3	Household	Meat, fish, and vegetable	Soy bean oil (65)
4	Korean charbroiling	Meat and seafood	Soy bean oil (23), Sesame oil (a little)

Notes: [a]The natural gas used in Beijing is from the northwest part of China in which methane, ethane, propane, butane and isobutene are major components.
[b]Salad oil mix means the restaurant uses diverse salad oil including rapeseed oil, soybean oil and peanut oil.

Table 2. The description of emission control facilities of four restaurants.

No.	Cooking style	Cooking condition	Air flow rate of ventilation ($m^3 h^{-1}$)	Seats	Dust catcher
1	Sichuan	6 stoves	12,000	170	Water spray and active charcoal adsorption
2	Zhejiang	22 stoves	44,000	400	High voltage static electric dust collector
3	Household	10 stoves	20,000	200	Active charcoal adsorption
4	Korean charbroiling	1 stove, and more than 10 charbroiling plates	12,000	352	Static electric dust collector

Table 3. Sampling description for each cooking style.

No.	Cooking style	Sampling time	Average smoke temperature (°C)	No. of samples
1	Sichuan	2 midday meals, 1 dinner	38	3
2	Zhejiang	2 midday meals, 1 dinner	42	3
3	Household	2 midday meals, 2 dinners	44	4
4	Korean charbroiling	2 midday meals, 1 dinner	42.5	3

2.2 *Sampling and hydrocarbon analysis*

The collection of cooking source samples were conducted by using 3.2 L fused silica lined (FLS) stainless steel canisters (Entech Instrument, Simi Valley, CA, USA), with a flow regulator (Entech CS 1200) that allows adjustment of the incoming flow at 34 mL min^{-1}. Thus, each sampling operation had lasted for 90 min, ensuring that the collected samples could be fully representative of the overall cooking activity. Emissions were sampled downstream from the exit of the ventilation exhaust pipeline using a Teflon pipe with a Teflon membrane filter to remove oil droplets and particles. Prior to sampling, pipes and connection parts were cleaned and checked for possible leakage, and canisters were cleaned by three cycles of pumping to vacuum, and flushing with high purity nitrogen. Samples were taken during both lunch and supper business time, and conducted every two days. The temperature of the oil fume was between 34 and 51°C while sampling. Ambient samples were taken simultaneously in a windward direction of the exit of ventilation exhaust pipeline. In total, 13 smoke samples and 13 atmospheric samples were obtained. Detailed sampling descriptions are shown in Table 3. Samples were sent to a laboratory at Peking University for immediate analysis. Hydrocarbon of these samples were firstly pre-concentrated through three cold traps (Model 7100, Entech Instruments, Simi Valley, USA), and then injected into a gas chromatography (GC, Model 6890, Hewlett Packard, USA) for separating using a semi-polar capillary column (DB624, 60 m × 0.32 mm i.d. × 1.8 μm). The oven temperature programme was −50°C for 3 min initially, to 164°C at 6°C min^{-1}, then to 200°C at 14°C min^{-1} and stable for 0.5 min finally. Separated hydrocarbons were detected by quadrupole mass spectrometer (MS, Model 5973, Hewlett Packard, USA). Bromochloromethane, 1,4-diflurobenzene and 1-bromo-3-flurobenzene were added into each sample as internal standard, and calibration curves were used to calculate the hydrocarbon concentration.

2.3 *Quality assessment/quality control*

Before sampling, 2–3 canisters were chosen at random from every batch of cleaned canisters and analysed to check the cleaning procedures.

For each batch of samples, 1-bromo-3-fluorobenzen tune was conducted to check the operating conditions of GC-MSD and to ensure the validity and reliability of results. Prior to analysing samples, daily calibration was performed to determine the median concentration in operating curve and to quantify the hydrocarbon samples. The deviation between daily calibration results and theoretical concentrations was within 20%. Blank tests were conducted to check the contaminant levels in the GC-MSD system. These tests ensured that there was no species detected in the chromatography signals.

Method detection limits (MDLs) were obtained by multiplying the standard deviation for seven duplicate measurements of 100 mL standard gas by 3.14 (the Student's t value for 99% confidence for seven data points). MDLs for various hydrocarbon species ranged from 0.009 ppbV to 0.040 ppbV in this study.

A group of ambient samples collected in parallel were done in duplicate, three to five times, and then the relative standard deviations (RSDs) were calculated for all target compounds to check the measurement precision. The average RSDs were within 15%.

3. Results and discussion

3.1 *Chemical speciation of hydrocarbon emissions*

The average total concentrations of hydrocarbons for Sichuan-style, Zhejiang-style, household-style and Korean barbecue were $466 \pm 3.8\,\mu g\,m^{-3}$, $601 \pm 25.8\,\mu g\,m^{-3}$, $463 \pm 84.9\,\mu g\,m^{-3}$ and $534 \pm 64.4\,\mu g\,m^{-3}$, respectively. This indicated that the hydrocarbon emissions from different cooking styles were comparative in this study. However, Sichuan-style and household-style emitted slightly less hydrocarbon than Zhejiang-style or Korean barbecue did. This was partly due to active charcoal adsorption used in the smoke treatments of the former two restaurants. The concentration of hydrocarbon from commercial cooking was roughly five to six times higher than the mean ambient concentration ($94 \pm 51.8\,\mu g\,m^{-3}$, $n = 13$). It is noteworthy that the hydrocarbons concentration in this study could not be the real HCs level in oil smoke directly emitted from cooking because all the samples had been treated by the cooking smoke control facilities. Thus, the HCs emission data in this paper may be lower than that under authentic emission conditions, and they are appropriate for regions with cooking smoke control requirements.

A total of 41 hydrocarbon species were quantified for the samples taken from cooking sources. For normalisation, the data were expressed as the mass percentage of the total hydrocarbon for each species (w%). Source profiles for all the selected cooking styles are listed in Table 4. The Chinese cooking source profile was the arithmetical mean of the profiles obtained from the three specific Chinese cooking styles.

All the restaurants had evident emissions of alkanes, alkenes and aromatics. Alkanes were the most abundant hydrocarbon group in the three Chinese restaurants, accounting for 37.9% in the total hydrocarbon mass concentrations, and the alkenes followed with 31.5%. In the emissions from the Korean charbroil restaurant, the alkenes were the most abundant group, which were about 10% higher than that of alkanes and aromatics. From a study on the hydrocarbon emissions from street vendor charbroiling of meat, the burning of charcoal did not emit alkenes [17]. In addition, meat barbecue using natural gas as fuel emitted abundant alkenes, while natural gas had minor composition of these unsaturated components [19]. Therefore, the high alkenes emission came probably from the high temperature treatment of meat.

The most abundant hydrocarbon species emitted from all the measured restaurants were consistent with each other. The top 10 hydrocarbon species with the highest concentrations from the cooking sources are shown in Figure 1. These species included 4 alkanes, 3 alkenes, and 2 aromatics, and totally accounted for 51.9% ~ 62.7% of the total hydrocarbon mass.

Table 4. Source profiles of hydrocarbon measured from four cooking sources (w%, average ± SD).

No.	Species	Sichuan style	Zhejiang style	Household style	Chinese cooking	Korean charbroiling
1	n-Butane	4.33 ± 1.28	3.28 ± 0.14	4.31 ± 1.18	4.06 ± 1.03	4.17 ± 0.80
2	Isobutane	14.44 ± 4.45	4.04 ± 0.12	3.18 ± 1.04	6.21 ± 5.41	7.53 ± 2.18
3	2-Methylbutane	2.67 ± 0.89	3.03 ± 0.25	1.96 ± 0.72	2.41 ± 0.77	4.16 ± 1.59
4	Pentane	4.67 ± 1.18	8.88 ± 0.65	14.3 ± 3.30	10.54 ± 4.86	2.94 ± 0.46
5	2,3-Dimethylbutane	0.38 ± 0.15	0.43 ± 0.06	0.23 ± 0.10	0.32 ± 0.13	0.29 ± 0.05
6	2-Methylpentane	1.77 ± 0.70	2.01 ± 0.36	1.00 ± 0.42	1.44 ± 0.63	1.38 ± 0.19
7	3-Methylpentane	1.59 ± 0.56	1.66 ± 0.43	0.77 ± 0.32	1.20 ± 0.57	1.20 ± 0.13
8	n-Hexane	2.71 ± 0.83	1.83 ± 0.20	1.80 ± 0.36	2.04 ± 0.57	1.28 ± 0.05
9	2,4-Dimethylpentane	0.80 ± 0.18	0.14 ± 0.02	1.61 ± 1.08	1.04 ± 0.97	0.05 ± 0.05
10	Methylcyclopentane	2.48 ± 1.24	0.97 ± 0.07	0.66 ± 0.21	1.19 ± 0.94	0.59 ± 0.04
11	Cyclohexane	0.55 ± 0.22	0.24 ± 0.01	0.20 ± 0.08	0.30 ± 0.18	0.15 ± 0.02
12	2,3-Dimethylpentane	1.36 ± 0.11	1.13 ± 0.27	0.73 ± 0.27	0.99 ± 0.35	0.80 ± 0.12
13	n-Heptane	0.98 ± 0.26	1.40 ± 0.03	3.11 ± 0.51	2.15 ± 1.10	0.80 ± 0.06
14	Methylcyclohexane	0.55 ± 0.09	0.54 ± 0.02	0.53 ± 0.22	0.54 ± 0.15	0.29 ± 0.04
15	Octane	0.81 ± 0.27	1.21 ± 0.08	3.29 ± 0.42	2.15 ± 1.26	0.65 ± 0.13
16	n-Nonane	0.45 ± 0.05	0.22 ± 0.20	0.46 ± 0.20	0.40 ± 0.19	0.27 ± 0.02
17	n-Decane	0.36 ± 0.00	0.31 ± 0.05	0.53 ± 0.08	0.43 ± 0.12	0.35 ± 0.26
18	Dodecane	0.67 ± 0.29	0.31 ± 0.06	0.55 ± 0.09	0.52 ± 0.19	0.41 ± 0.32
19	Propene	6.42 ± 2.95	6.42 ± 1.91	8.37 ± 0.74	7.39 ± 1.76	10.16 ± 1.78
20	trans-2-Butene	0.50 ± 0.03	1.63 ± 0.01	0.30 ± 0.11	0.68 ± 0.60	2.16 ± 1.01
21	cis-2-Butene	0.42 ± 0.05	1.08 ± 0.01	0.22 ± 0.10	0.49 ± 0.38	1.57 ± 0.77
22	1-Butene/Isobutene	2.25 ± 0.43	6.40 ± 0.85	4.43 ± 0.53	4.38 ± 1.65	7.80 ± 1.73
23	1-Pentene	1.08 ± 0.50	1.72 ± 0.30	2.21 ± 0.18	1.80 ± 0.56	2.67 ± 0.56
24	trans-2-Pentene	0.59 ± 0.05	0.99 ± 0.53	0.5 ± 0.12	0.65 ± 0.31	1.11 ± 0.10
25	2-Methyl-1-butene	0.77 ± 0.00	1.42 ± 0.56	0.62 ± 0.56	0.86 ± 0.55	1.04 ± 0.14
26	2-Methyl-2-butene	0.65 ± 0.12	1.70 ± 1.28	1.68 ± 3.36	1.43 ± 2.30	1.13 ± 0.26
27	2-Methyl-1-pentene/ 1-Hexene	1.18 ± 0.59	1.77 ± 0.49	2.63 ± 0.30	2.05 ± 0.74	3.07 ± 0.66
28	1-Heptene	1.19 ± 0.43	1.53 ± 0.50	2.94 ± 0.26	2.15 ± 0.90	2.34 ± 0.60
29	1-Octene	0.48 ± 0.17	0.70 ± 0.20	1.15 ± 0.13	0.87 ± 0.34	0.95 ± 0.24
30	trans-2-Octene	0.14 ± 0.20	0.39 ± 0.08	0.17 ± 0.34	0.22 ± 0.26	0.00 ± 0.00
31	Cyclopentene	0.21 ± 0.08	0.40 ± 0.00	0.35 ± 0.05	0.32 ± 0.09	0.62 ± 0.13
32	1,3-Butadiene	1.61 ± 0.38	2.26 ± 0.96	2.79 ± 0.19	2.36 ± 0.66	3.08 ± 0.17
33	Propyne	0.52 ± 0.14	1.65 ± 0.62	1.03 ± 0.16	1.06 ± 0.50	0.25 ± 0.07
34	Benzene	5.16 ± 1.16	10.25 ± 2.33	6.11 ± 0.86	6.91 ± 2.39	9.75 ± 2.99
35	Toluene	7.00 ± 0.30	8.41 ± 2.48	4.34 ± 1.34	6.03 ± 2.27	6.26 ± 0.57
36	Ethylbenzene	3.12 ± 1.39	1.88 ± 0.07	1.02 ± 0.37	1.76 ± 1.08	1.45 ± 0.13
37	m/p-Xylene	5.48 ± 3.24	1.86 ± 0.71	1.39 ± 0.80	2.53 ± 2.28	2.01 ± 0.11
38	o-Xylene	0.92 ± 0.08	1.26 ± 0.40	0.56 ± 0.30	0.83 ± 0.40	0.99 ± 0.07
39	3-Ethyltoluene	0.28 ± 0.06	0.64 ± 0.41	0.26 ± 0.11	0.36 ± 0.25	0.63 ± 0.50
40	1,2,4-Trimethylbenzene	0.58 ± 0.14	1.13 ± 0.51	0.55 ± 0.08	0.70 ± 0.33	0.98 ± 0.64
41	Styrene	0.38 ± 0.26	0.69 ± 0.13	0.32 ± 0.14	0.43 ± 0.22	1.15 ± 0.25
	Total	92.71	88.29	86.67	88.58	88.80

The major alkanes species were n-butane, iso-butane, 2-methyl butane and n-pentane. Although these species were commonly detected with higher concentration in cooking smoke, the specific percentages of them depended on cooking styles: as the most abundant species in Sichuan-style emission, the percentage of iso-butane was two to four times as high as that in emissions from other styles of cooking. The most abundant species from

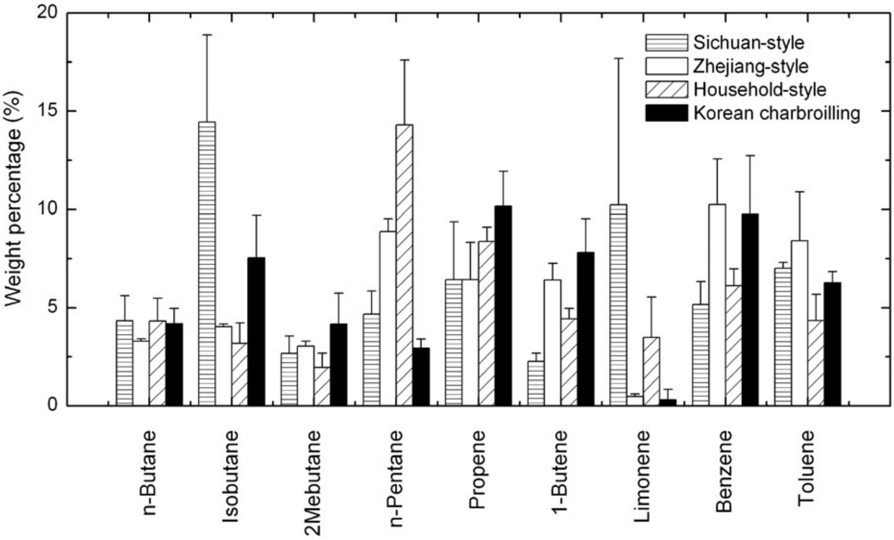

Figure 1. The top 10 hydrocarbon species measured from the emissions of the four restaurants.

household cooking was n-Pentane, contributing 14.3% to the total hydrocarbon. Without enough information, the sources of these alkane species could not be identified so far, but n-butane and iso-butane could be partly from incomplete combustion of natural gas.

Propene, 1-butene, and iso-butene were the alkene species that were commonly identified in the cooking emissions, with the sum of the three accounting for about 40% in the total hydrocarbon mass. A significant amount of limonene was found from emissions of Sichuan-style and household-style restaurants, which accounted for about 35% and 11% respectively of the alkenes mass, while the limonene from Zhejiang-style restaurants and Korean charbroil were almost around the detection limit of our GC-MS system. Limonene emitted from the flavouring materials and some vegetables [25–27]. In the Sichuan- and household-style cooking practices, large amounts of Sichuan pepper were used to obtain the unique Sichuan spicy taste, and the volatile matter from Sichuan pepper had a content of limonene as high as 20% [26]; the other styles of cooking activities did not use this kind of flavouring ingredient.

In cooking emissions, the predominant aromatics were toluene and benzene, which accounted for 7.7% and 6.1% respectively of total hydrocarbon. Besides these two species, high percentages of m/p-xylene and ethylbenzene were found in the Sichuan-style restaurant emissions, as 5.5% and 3.1% respectively of the total, but they were relatively low, less than 2%, in the emissions from other three restaurants.

The hydrocarbon emissions were determined by the procedure of various cooking activities, relating to the raw materials, oils and fuels used for cooking. For example, the aromatics were investigated to be from cooked meat and oil [20,28], and also from incomplete burning of wood [17,21]. Generally, hydrocarbon was mainly from raw materials and cooking oil, the fuels had a minor contribution [12,17,21]. In this study, because the hydrocarbon emission profiles were much more complex than the chemical speciation of natural gas, we considered that the main hydrocarbons identified were largely from cooking processes of raw materials and oil, not from fuel. Furthermore, the cooking style and the cooking fume treatment technologies could also affect the hydrocarbon emissions. So, further studies need to be done for identification of impact factors and estimation of their effect.

The source profile uncertainties could not be calculated as relative standard deviation of duplicate samples due to limited samples. Generally, the analytical uncertainties were in the range of ±10–15% for weighted percentage exceeding five times of the minimum detected limit (MDL). The default coefficient of variability of ±20% was assigned to all species whose weight percentage was larger than 0.1%. For those less than 0.1%, the uncertainty was estimated as the following formula:

$$\sigma = \sqrt{MDL^2 + (CV \cdot \omega t\%)^2}$$

where CV is the coefficient of variation and $\omega t\%$ is the weighted percentage of various species in the source profile [29]. Because of the variability among individual restaurants or range of cooking operating conditions, the source profile uncertainty for hydrocarbon species from cooking in this paper can exceed the variability due to measurement error. Thus, to acquire representative source profiles for each cooking style, more restaurants of the same style and representative cooking operating conditions should be studied in further work.

3.2 *Comparison of hydrocarbon emissions from different styles of cooking*

The various tastes of Chinese cuisine come from localised recipes of raw materials and special cooking practices, and then in turn result in different emissions of hydrocarbon. In others' work, four kinds of Mexican cuisines were used to build correspondent source profiles for hydrocarbons [21], and analysis on organic emissions from typical cooking activities such as barbecue of meat and frying of vegetables was performed in the United States [19–20]. In this study, we did the measurement for four kinds of restaurants in China, deploying an identical sampling method as that used in the above studies. Therefore, the source profiles from our work and from the literature could be compared together for better understanding of hydrocarbon emissions features.

3.2.1 *Chinese cooking versus Mexican and American cooking*

Although the hydrocarbon species emitted from Chinese restaurants differed slightly, the three styles of restaurants (Sichuan, Zhejiang and household style) had quite similar hydrocarbon compositions in our measurements. Therefore, the average hydrocarbon compositions of these three restaurants were used as the hydrocarbon source profile of Chinese cooking. In the studies on food preparation of Mexico City, popular Mexican dishes cooked in restaurants were incorporated to compare. We noted that frying meat and pigskin with pork lard was an authentic Mexican style and very similar with Chinese cooking in terms of raw material and oils. Thus, frying food with pork lard was selected for comparison as well. In the study on fried vegetables of United States, the hydrocarbon emissions from vegetable fried in soybean oil were also used for comparison.

Prior to comparison, the hydrocarbon species that were commonly quantified were selected from different studies, and the relative abundances of hydrocarbon species were expressed as the mass percentages of a certain species in the sum of these common species (Figure 2a and b).

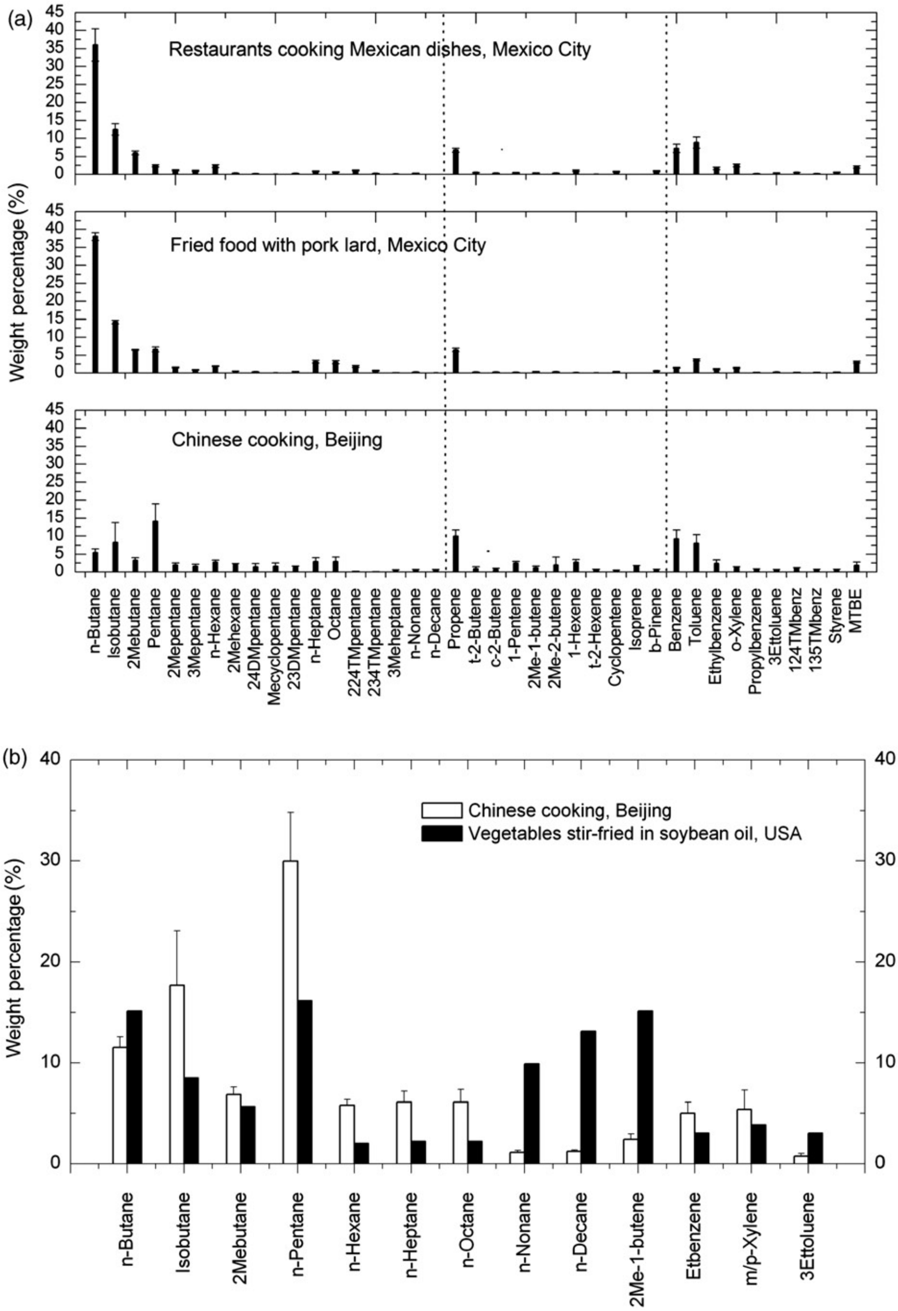

Figure 2. The comparison of hydrocarbon profiles from Mexican-style and Chinese-style cooking: (a) comparison between Mexican normal-style cooking, the special fried food with pork lard, and Chinese cooking; (b) vegetables fried in soybean oil.

The normal Mexican-style cooking and the special fried food with pork lard were compared with Chinese cooking in Figure 2a. These profiles had similarity in the major hydrocarbon species, namely lower carbon number like alkanes, propene, benzene and toluene. But the two hydrocarbon profiles of Mexican cooking showed results as high as 35% of n-butane, compared with only 5% measured in Chinese cooking oil fumes. Iso-butane had also a much higher percentage in Mexican cooking than that of Chinese cooking. This difference might be very likely due to the difference in cooking fuels, because the two Mexican restaurants used liquefied petroleum gas (LPG) that consisted of predominantly n-butane and iso-butane, while the Chinese restaurants used mainly natural gas, with methane as the major content. The aromatics had very similar percentages for the normal Mexican cooking and Chinese cooking, but the fried food with pork lard showed evidently lower aromatics content.

In the emissions from the vegetables fried in soybean oil, only 13 hydrocarbon species were commonly identified, as shown in Figure 2b. C4–C5 alkanes were the major components from both Chinese and American cooking processes. Different from Chinese cooking, the American frying vegetable emitted much higher percentages of large alkanes like nonane and decane, and also 2-methyl-1-butene. As the fuel and cooking oil were the same, we attributed this difference to the difference in recipe of raw materials.

3.2.2 *Comparison of Barbecue emissions*

Barbecue is very popular over the world, and also in vogue in the cities of China. This kind of cooking uses meat as the main material and with very little oil, thus it was convenient for comparison. In this study, comparison among different barbecue source profiles was done, including Korean-style charbroiling, American barbecue using natural gas as fuel [19], and Mexican rotisserie [21]. Chinese cooking was also added for comparison. Finally, a total of 20 hydrocarbon species were selected for follow-up discussion.

As shown in Figure 3, the main hydrocarbon species from barbecue sources were C4-C5 alkanes, propene, benzene and toluene. Due to incomplete combustion of LPG fuel, Mexican barbecue showed very high abundant n-butane and iso-butane, which accounted for more than 50% of the total 20 species mass. Korean-style charbroiling of this study released higher percentages of aromatics than other barbecue sources, which could partly come from burning of charcoal [21]. Interestingly, the American style of barbecue had higher emissions of unsaturated species, e.g. up to 30% of the total hydrocarbon was propene. This was also the case for Korean-style charbroiling, but slightly lower than American-style barbecue. Chinese cooking had a very different species distribution in alkane but a similar pattern of aromatics compared with barbecue sources.

3.3 *Hydrocarbon emission factors*

The hydrocarbon emissions from cooking source were influenced by raw material, oil and fuel, so it was difficult to calculate the emission factors taking all these parameters into consideration. From previous studies, we considered that the cooked raw materials could be the major parameter, and then performed a survey in the Sichuan-style restaurants for the usage of meat, and calculated the hydrocarbon emission factors based on the consumed meat (Table 5). In the Sichuan-style cooking activities, the hydrocarbon emission factor was measured to be 0.29 g per kg of meat cooked, and this emission factor was much lower than barbecue sources [17,19], but about 40 times as high as that from

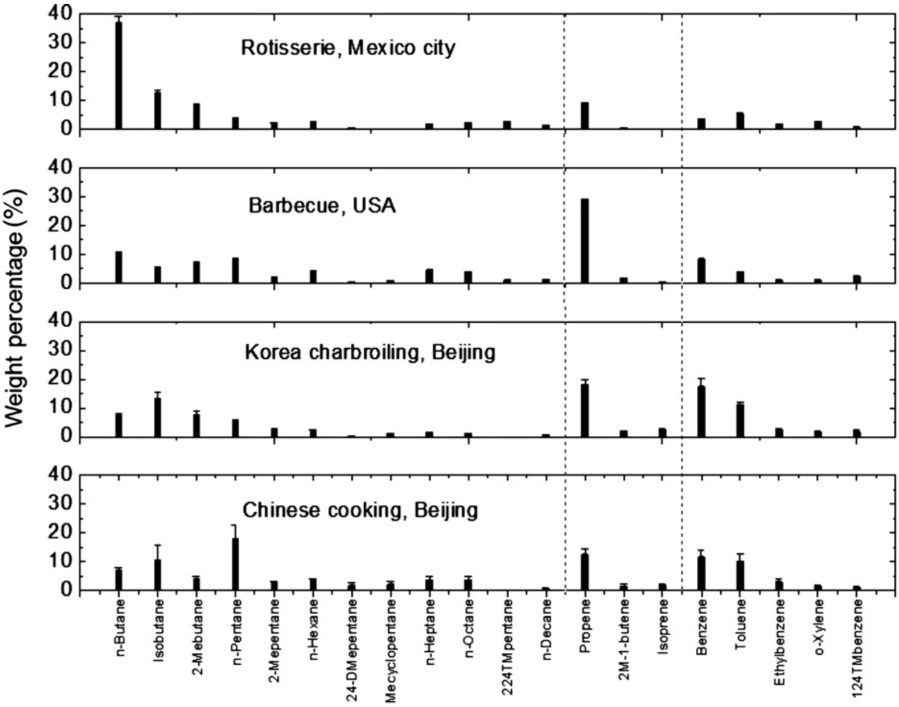

Figure 3. The comparison of hydrocarbon source profiles of Korean, Mexican and American style of barbecue and Chinese cooking.

Table 5. Emission factors of hydrocarbon and some hydrocarbon species from several cooking sources (mg kg^{-1} meat cooked).

Species	Sichuan style	Meat charbroiling	Vegetables fried in soybean oil	Charcoal grilling
n-Butane	12.7	107	0.75	
Iso-butane	42.4	54.0	0.42	
2-Methylbutane	7.83	74.0	0.28	
n-Pentane	13.7	87.0	0.80	
n-Hexane	7.94	44.0	0.10	
Methylcyclopentane	7.26	9.0		
Propene	18.8	289		
1-Butene	6.61	73.0		
Limonene	30.0			
Benzene	15.1	83.0		466
Toluene	20.5	40.0		179
Ethylbenzene	9.14	11.0	0.15	34.5
m/p-Xylene	16.1	12.0	0.19	25.0
Sum of hydrocarbon	*293*	*1205*	*4.95*	*4940*
Reference	This study	[19]	[20]	[17]

vegetables fried with soybean oil [20]. The emission factors of some typical hydrocarbon species were also given in Table 5 according to obtained source profiles.

Although limited samples (only three) had been done in the Sichuan-style, the emission factor values in this study are reliable due to using a sound analytical method and taking duplicate tests, according to rating criterion of AP 42 Compilation of Air Pollutant Emission Factors (AP-42) released by US EPA.

Rough estimation of contribution to total VOC emission of Beijing was done based on a synthesis of Xie *et al*. [30]. The hydrocarbon emissions from commercial cooking restaurants registered accounted for about 1% of total VOC emissions in 2008 in Beijing. But this data was excessively underestimated because not all the VOC species were included, and thousands of restaurants that were not registered, street vendors and domestic cooking operations were excluded here.

4. Conclusions

Emissions from the cooking process of commercial restaurants were measured in Beijing to obtain hydrocarbon source profiles and for further application for hydrocarbon source apportionment. This study measured four cooking styles in China, including Sichuan cuisine, Zhejiang cuisine, household-style cuisine and Korean-style barbecue. All the selected restaurants emitted a comparable amount of alkanes, alkenes and aromatics, with n-butane, iso-butane, 2-Methylbutane, n-pentane, propene, 1-butene, benzene, and toluene as the most abundant species. Sichuan-style and household cooking emitted a high proportion of limonene. There were quite large differences in hydrocarbon source profiles with Chinese cooking in this study and results from other cities in the world, perhaps due to the use of raw material and fuels for cooking. Comparing emissions of barbecue from different regions it was found that they were all impacted by fuel compositions significantly. Hydrocarbon source profiles of barbecue emissions differed from those of Chinese-style cooking. Emission factors of various VOC species emitted from Sichuan-style restaurants were calculated, and found to be much lower than that from meat charbroiling.

The profiles obtained in this paper can be used in the CMB model together with hydrocarbon profiles from other sources. However, more cooking styles should be included in a further study, in order to improve representation and reduce the uncertainty of source appointment.

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