

# Composition profiles of organic aerosols from Chinese residential cooking: case study in urban Guangzhou, south China

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**Abstract** Residential cooking in China's urban areas could be a more significant contributor to organic aerosols when compared to restaurant cooking, yet no source profiles are available for Chinese residential cooking. In this study, a typical nine-floor residential apartment building in urban Guangzhou was selected to investigate the particulate mass emissions and chemical compositions of organic aerosols from Chinese residential cooking. During dinner cooking period, the average of total suspended particle (TSP) mass concentrations increased by a factor of 4.40 compared with that during non-cooking period. Organic matter (OM) was the major component of aerosols from cooking, and accounted for 66.9 % of the TSP mass. Over 90 organic species were identified and quantified, and could explain 14.5 % of OC in cooking emissions. Fatty acids alone shared 75.7 % of the total mass of quantified organic compounds, with palmitic acid as the predominant one. Sterols, monosaccharide anhydrides and polyols were also significant compositions and they together contributed 14.3 % in the total mass of the quantified organic compounds. While cholesterol was detected from western-style cooking, phytosterols were additionally observed from Chinese-style cooking, and campesterol and polyols were only reported in the present study. Chinese residential cooking emission showed small differences in composition patterns of organic compounds when compared to previously report Chinese restaurant cooking emissions, however, large differences existed when compared to western style cooking. Using cholesterol as a tracer for cooking emissions, in Guangzhou we found cooking emissions contributed 3.9–8.2 % of OC in PM<sub>2.5</sub>. Spatial distributions of cooking emission contribution were consistent with the distributions of population.

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## 1 Introduction

Many studies have indicated a positive relationship between fine particulate matter (PM<sub>2.5</sub>) and human health endpoints (Pope et al. 2002; Deng et al. 2008; Xu et al. 2008; Shang et al. 2013). Organic matters (OM) are major components of PM<sub>2.5</sub> and can contribute up to 70 % of the PM<sub>2.5</sub> mass (Turpin et al. 2000; Zheng et al. 2005). However, sources apportioning of particulate OM is still a challenge since these organics can come from various primary emission sources and as well from secondary formation in the atmosphere. One of the most important but uncontrolled sources in urban areas is cooking emission (Hildemann et al. 1991; Wang et al. 2009; He et al. 2010), which is predominantly carbonaceous in its chemical compositions (See and Balasubramanian 2008), with air toxics like polycyclic aromatic hydrocarbons (PAHs) in the fume (Zhu and Wang 2003). Previous studies have revealed that cooking emission accounted for 10–30 % of organic carbon (OC) in megacities around the world (Schauer et al. 1996; Robinson et al. 2006; Zheng et al. 2006; Wang et al. 2009).

A big problem to decipher the contribution of cooking emission is that its source profiles vary greatly among different cooking styles (He et al. 2004; Zhao et al. 2007a; Hu et al. 2010) or different cooking foods, such as meat charbroiling (Rogge et al. 1991; Schauer et al. 1999; McDonald et al. 2003) and vegetables cooking with seed oils (Schauer et al. 2002). As a result, Robinson et al. (2006) found that, using different profiles, contributions of cooking emission to OC in ambient PM<sub>2.5</sub> estimated by chemical mass balance (CMB) exhibited a difference by a factor of more than 9. So it is of vital importance to obtain the representative source profiles for cooking emission when investigating its contribution.

Cities in China are now facing severe fine particle pollution, with ambient PM<sub>2.5</sub> levels frequently beyond 100 µg/m<sup>3</sup> (He et al. 2001; Wang et al. 2006) and POM as the most abundant chemical components (Wang et al. 2012). In China, urban areas typically mixed with residential/commercial/administration areas are much more densely populated than rural areas. For example, some areas in urban Guangzhou, a megacity in south China, have population density of over 30,000 permanent residents per square kilometer. Moreover, Chinese-style cooking has more frying than the western-style ones, and thus emits larger amounts of organics emitted due to high-temperature processing of foods (Zhao et al. 2007a, b). Considering the population density and the characteristics of Chinese-style cooking, cooking emissions are probably a source of great importance particularly in China's urban areas.

Majority of residents in China's urban areas dwell in multi-story apartment buildings, e.g., 89 % of residents are apartment-dwellers in Guangdong province, south China (Guangzhou Statistical Yearbook, 2012). Cooking emissions from restaurants in the urban areas of China's large cities are required to be further treated to, at least, avoid complaints from residents in the neighborhood. However, in multi-story apartment buildings, cooking fumes in each kitchen are sucked by a range hood and introduced directly to the atmosphere by a shared chimney or exhaust pipe without any treatment except that the range hoods can separate oils from the smoke and get them collected in appropriate containers. Considering the fact that most people cook their meals at home, we could expect substantial contribution of cooking emission from residential buildings. However previous investigations in Chinese cooking emission were mostly targeted on emission from restaurants of different styles (He et al. 2004; Zhao et al. 2007a; Hu et al. 2010). To the best of our knowledge, there are no source profiles available for Chinese residential cooking emission.

In the present study, we collected aerosol samples in the outlet of a shared chimney on a typical multi-story apartment building and get them analyzed for organic species. The objectives are (1) to determine the organic compositions and source profiles of Chinese residential cooking; (2) to get a tracer-based estimate for the contribution of cooking emission to organic aerosols with chemical composition data at typical urban, suburban and rural sites in Guangzhou.

## 2 Experimental

### 2.1 Sampling

A typical nine-floor residential multi-story apartment building, with four apartments in each story, in the Guangzhou Institute of Geochemistry, Chinese Academy of Sciences (GIG) in urban Guangzhou (Fig. 1) was selected to investigate the source profiles of Chinese residential cooking emissions in this study. Among the four apartments in each story, two are 85 m<sup>3</sup> (inside area) ones, with one living room, three bed rooms, one kitchen and one toilet in each one; two are 97 m<sup>2</sup> ones with one living room, four bed rooms, one kitchen and one toilet in each one. All the apartments are occupied, and each apartment hosts a family. The residents in the building were born and raised in nine provinces of China. Their cooking styles cover major ones in China, including Guangdong-, Sichuan-, Shandong-, Jiangsu-, Zhejiang- and Fujian-styles. Source profiles of the cooking emission from the shared chimney would be more representative of cooking emission in urban areas due to mixed cooking styles of residents inside the apartment building. Pipeline natural gas is used as the cooking fuel for all apartments. As showed in Fig. 2, in each kitchen an overhead range hood is placed on the cooking stove to withdraw the cooking fumes to the shared exhaust pipe with the outlet on the roof of the building. Two apartments of each story shared one exhaust pipe.

During a campaign from August 21 to September 5, 2012, we collected filter-based TSP samples as well as filed blanks at a flow rate of about 10 L/min by placing the sampling probe of an emission source sampling system (Isokinetic source sampler, Model 500-Series) directly into the outlet of the exhaust pipe on the roof of the building. Questionnaire survey among the residents in the building demonstrated that 91.7 % of the families cook their dinners at home, while only 58.3 % of the families cook their lunches at home (Table S1). So we collected aerosol samples at the pipe outlet during the dinner cooking time interval of 17:00–20:00 in the evening to represent the cooking exhausts and also during the time interval of 14:00–17:00 in the afternoon without cooking activities for contrast. Before sampling, the filter holders and open ends of the samplers were all cleaned with ethanol and immediately wrapped by clean aluminum foil until field sampling. During sampling quartz filters (82.6 mm diameter, Whatman), which were prebaked at 450 °C for 4 h to remove any organic contaminants, were loaded onto the sampler holders to collect the aerosols samples. After sampling, samples were wrapped by clean aluminum foil and stored in a refrigerator at –20 °C until lab analysis.

To get tracer-based estimates for the contribution of cooking emissions to OC in Guangzhou, we also collected PM<sub>2.5</sub> samples for the chemical compositions at typical sampling sites in Guangzhou. Locations of these sampling sites were depicted in Fig. 1 and the description of the sampling sites and field sampling were detailed in our previous paper (Zhao et al. 2014).

### 2.2 Laboratory analysis

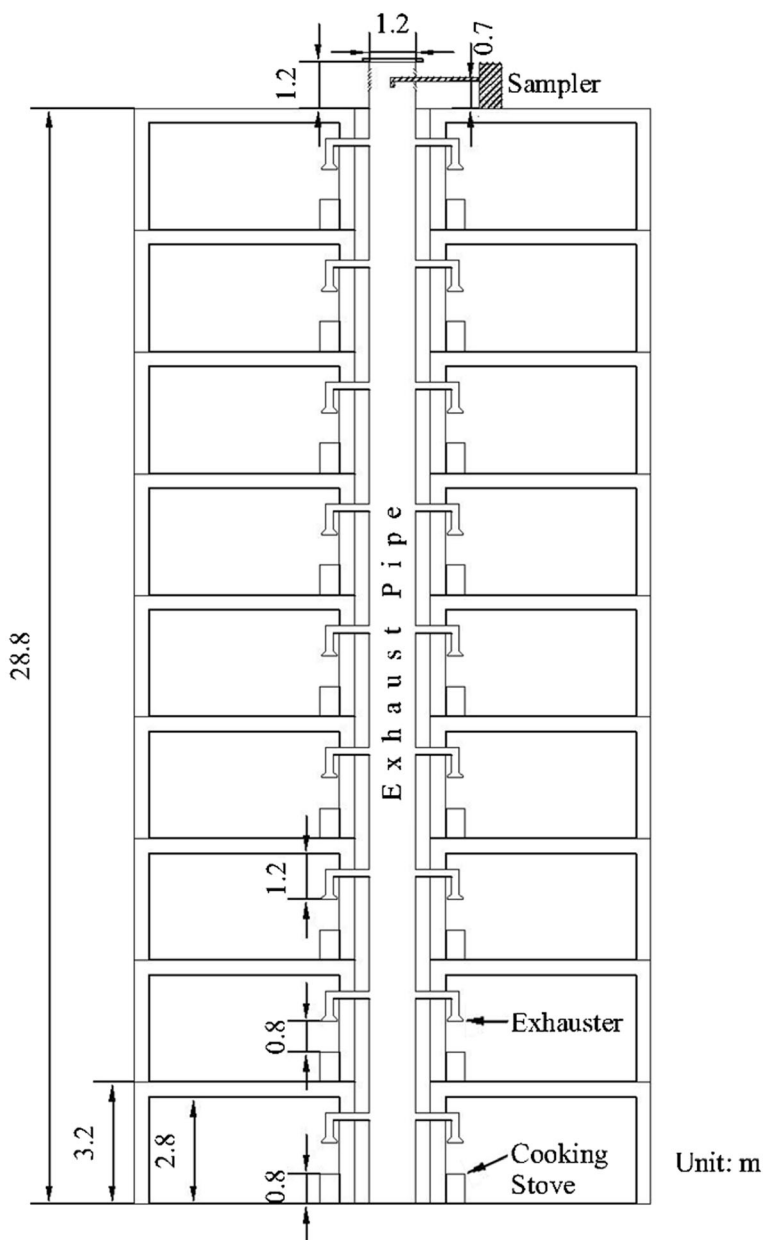
Particulate mass concentrations were determined by weighing the filters before and after the sampling in a separated weighing room. A punch (1.5×1.0 cm<sup>2</sup>) of each filter was taken and



**Fig. 1** Sampling sites in Guangzhou, south China

analyzed for organic carbon/elemental carbon (OC/EC) by an OC/EC Analyzer (Sunset Laboratory Inc., USA) with the thermo-optical transmittance (TOT) method (NIOSH 1999).

Another punch of each filter was taken and analyzed for solvent-extractable organic compounds. The analytical procedures in this study were similar with those described by Ding et al. (2011). 1/2 of each filter was first spiked with 100  $\mu\text{L}$  internal standards including n-tetracosane- $\text{D}_{50}$ , lauric acid- $\text{D}_{23}$ , palmitic acid- $\text{D}_{31}$ , phthalic acid- $\text{D}_4$ , levoglucosan- $^{13}\text{C}_6$ , and ultrasonically extracted twice with 30 mL hexane/dichloride methane (DCM) (1:1, v/v) each time and then twice with 30 mL DCM/methanol (1:1, v/v) each time. The extracts were combined, filtered and concentrated to about 1 mL. Then each of the concentrated extract was split into two parts. One part was methylated with fresh prepared diazomethane to convert organic acids to methyl esters. The methylated extract was analyzed for alkanes, fatty acids,  $\text{C}_4$ - $\text{C}_9$  dicarboxylic acids, aromatic acids, polycyclic aromatic hydrocarbons (PAHs) and poly acids. The other part was silylized with 25  $\mu\text{L}$  pyridine and 50  $\mu\text{L}$  N, O-bis-(trimethylsilyl)-trifluoroacetamide (BSTFA) plus 1 % trimethylchlorosilane (TMCS) in an oven at 70  $^\circ\text{C}$  for 1 h. The silylated extract was analyzed for polyols, sterols and levoglucosan. Both derivatized fractions were analyzed using Agilent Model 5975 N gas chromatography-mass selective detector (GC-MSD) equipped with an Agilent HP-5 MS capillary column (30 m in length  $\times$  0.25 mm I.D.  $\times$  0.25  $\mu\text{m}$  film thickness). The GC temperature was initiated at 65  $^\circ\text{C}$  and held for 2 min. Then the temperature was increased to 290  $^\circ\text{C}$  at a rate of 5  $^\circ\text{C}/\text{min}$  and held for 20 min. Most of organic compounds were quantified by authentic standards, the other compounds which lack of standards was quantified using adjacent similar compounds (Table S2).



**Fig. 2** Schematic diagram of the sampling in the residential apartment building

### 2.3 Quality assurance/quality control (QA/QC)

Recoveries of the target compounds in six spiked samples (authentic standards spiked into solvent with pre-baked quartz filter) were ranged from 66.7 to 128 %. The field blanks were extracted and analyzed in the same way as the field samples. The reported results were corrected for the field blanks, but not recovery corrected.

### 3 Results and discussion

#### 3.1 Particle mass concentration

The filter-based TSP increased from an average level of  $69.9 \mu\text{g}/\text{m}^3$  during the afternoon non-cooking period (14:00–17:00) to that of  $208 \mu\text{g}/\text{m}^3$  during the evening dinner cooking period (17:00–20:00) (Table S3). To eliminate the influence of background ambient air, we subtracted concentrations of TSP mass and speciated organics in afternoon samples from those in evening samples. Thus the average concentration of TSP from the Chinese residential cooking was  $138 \mu\text{g}/\text{m}^3$ . As shown in Table 1, big differences were observed among different cooking styles due to combined influences of many factors, such as cooking styles, materials, temperature, fuel styles and also sampling conditions and monitoring methods. Compared with  $\text{PM}_{2.5}$  concentrations monitored by Wan et al. (2011) in a residential kitchen during cooking, the present results of TSP were about  $30 \mu\text{g}/\text{m}^3$  lower, suggesting obvious adsorption of particles by the exhaust system. Generally, particle mass concentrations monitored in rooftop exhausts from Chinese residential cooking were about one magnitude lower than those from restaurant cooking (He et al. 2004), indicating less particle emissions from residential cooking. In addition, OM (calculated as 1.4 times of OC) (Zhao et al. 2007a) accounted for 66.9 % of TSP mass from Chinese residential cooking.

#### 3.2 Organic composition of residential cooking emissions

The average concentration of OC during cooking period ( $85.4 \mu\text{g}/\text{m}^3$ ) was 3 times higher than the background levels during non-cooking periods ( $19.4 \mu\text{g}/\text{m}^3$ ). Over 90 species of organic compounds were identified and quantified. Detected organic compounds, such as fatty acids, sterols, levoglucosan and polyols, also showed 3–12 times higher concentrations during the evening cooking period than those during non-cooking period (Table S3).

All the organic compounds were normalized to OC for comparison with results from previous studies. The ratios of specific organic compounds to OC (ng/mg OC) from cooking in this study were shown in Table 2. The quantified organic compounds accounted for 9.50 % of OC in the residential cooking emissions. This percentage was within the range of 7.21–32.6 % in Chinese restaurant cooking emissions (He et al. 2004; Zhao et al. 2007a; Hu et al. 2010). Quantified organics were reported to account for less than 4 % of OC in meat cooking emissions (Hildemann et al. 1991; Rogge et al. 1991), but over 80 % of OC from seed oils cooking and western-style fast food cooking which emitted substantial fatty acids (Schauer et al. 2002; Zhao et al. 2007b). However, meat cooking might produce massive complicated high molecular compounds like heterocyclic amines that could not be detected by the GC-MSD method (Thiebaud et al. 1995).

As shown in Fig. 3, saturated and unsaturated fatty acids were the most abundant organic compounds from the residential cooking emissions, and accounted for 75.7 % of the total quantified organic compounds. Sterols, monosaccharide anhydrides and polyols were also significant compositions, and together contributed to 14.3 % of the total mass of quantified organic compounds. Nevertheless, dicarboxylic acids, alkanes, PAHs, poly acids and aromatic acids were only minor fractions.

##### 3.2.1 Fatty acids

$\text{C}_4$  to  $\text{C}_{34}$  even carbon number fatty acids are main constituents of triglycerides and phospholipids in vegetable oils and animal fats, with  $\text{C}_{16}$  and  $\text{C}_{18}$  fatty acids as major species (Rogge

**Table 1** Particle mass concentrations measured during cooking activities ( $\mu\text{g}/\text{m}^3$ )

Experimental condition	Size	Conc.	References
Real cooking (rooftop exhausts)			
Chinese residential	TSP	138	This study
Chinese restaurants, Hunan	PM2.5	1406	(He et al., 2004)
Chinese restaurants, Cantonese	PM2.5	672	
Real cooking (kitchen)			
Chinese residential	PM2.5	160	(Wan et al. 2011)
Chinese commercial	PM2.5	312.4	(See and Balasubramanian 2006)
Chinese stall	PM2.5	201.8	(See et al. 2006)
Malay stall	PM2.5	245.3	
Indian stall	PM2.5	186.9	
Chinese hot-pot restaurant	PM2.5	81.1	(Lee et al. 2001)
Chinese dim sum restaurant	PM2.5	28.7	
Western Canteen	PM2.5	21.8	
Korean barbecuing restaurant	PM2.5	1167	
Pizzerias	PM1	10–327	(Buonanno et al. 2010)
	PM2.5	12–368	
	PM10	15–482	
Controlled cooking (kitchen)			
Steaming	PM2.5	65.7	(See and Balasubramanian 2008)
Boiling	PM2.5	81.4	
Stir-frying	PM2.5	120	
Pan-frying	PM2.5	130	
Deep-frying	PM2.5	190	
Grilling in a gas stove at maximum power with:			(Buonanno et al. 2009)
Cheese	TSP	283	
Wurstel sausage	TSP	352	
Bacon	TSP	389	
Eggplant	TSP	78	
Frying 50 g of chips in a gas stove at maximum power with:			
Olive oil	TSP	118	
Peanut Oil	TSP	68	
Sunflower Oil	TSP	60	
Frying 50 g of chips using an electrical pan with:			
Olive Oil	TSP	27	
Peanut Oil	TSP	13	
Sunflower oil	TSP	12	
Tofu boiling	PM2.5	22.88	(Huboyo et al. 2011)
Tofu frying	PM2.5	41.12	
Chicken boiling	PM2.5	30.86	
Chicken frying	PM2.5	101.64	
Lab simulate domestic kitchen:			(To and Yeung 2011)
Frying vermicelli with beef using gas cooking	PM10	1330	

**Table 1** (continued)

Experimental condition	Size	Conc.	References
Frying vermicelli with beef using electric cooking	PM10	1030	
Pan Frying of meat using gas cooking	PM10	1020	
Pan Frying of meat using electric cooking	PM10	520	
Deep frying of chicken wings using gas cooking	PM10	890	
Deep frying of chicken wings using electric cooking	PM10	680	
Lab simulate commercial kitchen:			
Deep frying of tofu using gas cooking	PM10	4720	
Deep frying of tofu using electric cooking	PM10	3980	
Griddle frying of meat using gas cooking	PM10	2260	
Griddle frying of meat using electric cooking	PM10	2600	
Broiling of beef satay using gas cooking	PM10	out of range	
Broiling of beef satay using electric cooking	PM10	out of range	

et al. 1991). Typically, vegetable oils have more unsaturated fatty acids than animal fats (Zhu 1995). During the cooking process, free fatty acids are released by hydrolysis and thermal oxidation of glycerides (Rogge et al. 1991). In this study, C<sub>9</sub> to C<sub>28</sub> fatty acids from cooking emissions were determined, and the concentrations were listed in Table 2.

Saturated fatty acids emitted from residential cooking had an average total concentration of 56,508 ng/mg OC, which is in the same order of magnitude with 22,911–126,976 ng/mg OC reported previously from Chinese restaurant cooking in Guangzhou (Zhao et al. 2007a), Shenzhen (He et al. 2004) and Hong Kong (Hu et al. 2010). Saturated fatty acids emitted from meat cooking were found to be in low levels of 1078–49,548 ng/mg OC according to studies by Rogge et al. (1991) and Schauer et al. (1999). However, seed oil cooking (Schauer et al. 2002) and the western-style fast cooking (Zhao et al. 2007b) emitted about 10 times of saturated fatty acids (238,974–524,579 ng/mg OC), which was attributed to ingredients and high cooking temperature. C<sub>9</sub>–C<sub>28</sub> saturated fatty acids accounted for 48.6 % of total quantified organics. The compositions were characterized with significant even-to-odd carbon preference and palmitic acid as the most abundant compound, which agreed well with results from previous studies (Rogge et al. 1991; Schauer et al. 1999, 2002; He et al. 2004; Zhao et al. 2007a, b; Hu et al. 2010). Besides, nonanoic acid presented a peak level among C<sub>9</sub>–C<sub>15</sub> odd carbon number saturated fatty acids.

Unsaturated fatty acids accounted for 27.1 % of total quantified organic compounds in the present study, in which oleic acid and linoleic acid were the most abundant species. This percent was within the range of 7.41–62.1 % in previous cooking emission studies (Rogge et al. 1991; Schauer et al. 1999, 2002; He et al. 2004; Zhao et al. 2007a, b; Hu et al. 2010).

As shown in Fig. 4, dominant fatty acids differed among emissions from different styles of cooking. Palmitic acid peaked in residential and Sichuan- and Dongbei-style restaurant cooking emissions, while oleic acid or linoleic acid were the highest in Canton- or Hunan-style restaurant cooking emission and seed oil cooking emissions. The level of oleic acid, an organic molecular marker widely used to estimate the contribution of cooking emissions, was found to be 1.3–5 times lower in the emissions from residential cooking than those from Chinese restaurant cooking (He et al. 2004; Zhao et al. 2007a; Hu et al. 2010). It is suggested that there would be large uncertainties when assessing the contribution of cooking emissions to ambient aerosols using this marker due to great variations on emission profiles.



**Table 2** Mass fractions of each organic compound in particles from the Chinese residential cooking (ng/mg OC). The data has been corrected for background ambient air

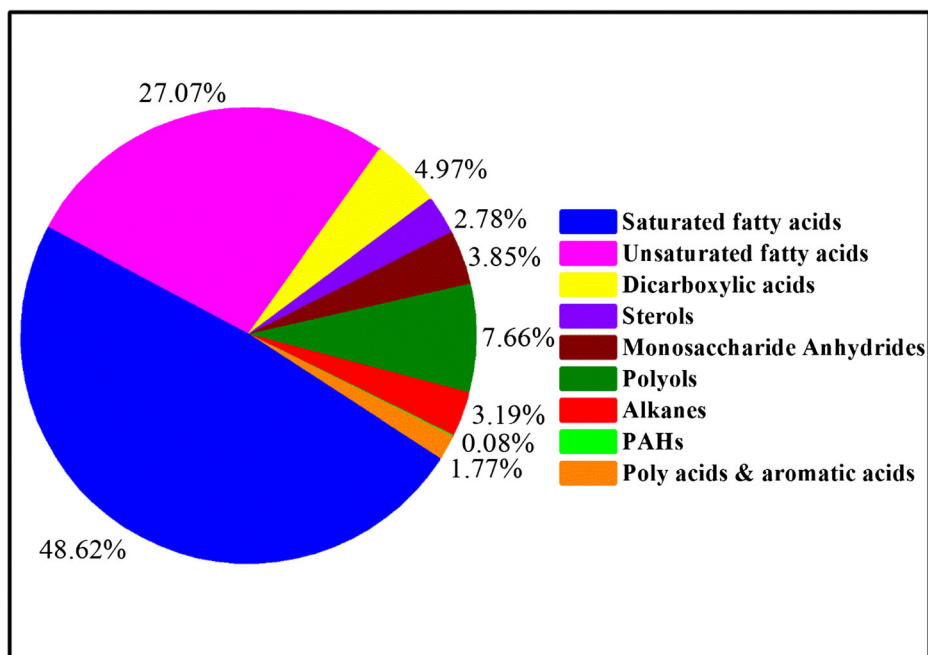
Target Compounds		Target Compounds	
Fatty acids			
Saturated fatty acids			
Nonanoic acid	5156±530	Nonadecanoic acid	44.3±2.73
Decanoic acid	45.6±27.6	Eicosanoic acid	977±83.2
Undecanoic acid	15.9±3.95	Heneicosanoic acid	57.9±2.56
Dodecanoic acid	281±31.3	Docosanoic acid	976±88.7
Tridecanoic acid	55.5±5.27	Tricosanoic acid	60.5±3.22
Tetradecanoic acid	1269±75.0	Tetracosanoic acid	302±23.8
Pentadecanoic acid	419±27.9	Pentacosanoic acid	19.1±1.33
Hexadecanoic acid (Palmitic acid)	34301±2458	Hexacosanoic acid	46.0±3.10
Heptadecanoic acid	465±30.3	Heptacosanoic acid	4.17±0.52
Octadecanoic acid (Stearic acid)	11,993±993	Octacosanoic acid	20.8±2.16
Unsaturated fatty acids			
9-Hexadecenoic acid (Palmitoleic acid)	739±102	9-Octadecenoic acid (Oleic acid)	14,905±1489
9,12-Octadecanedienoic acid (Linoleic acid)	15,821±1611		
Dicarboxylic acids			
Adipic acid	200±20.4	Azelaic acid	4177±411
Pimelic acid	360±31.6	Sebacic acid	223±17.6
Suberic acid	819±67.1		
Sterols			
Cholesterol	785±67.3	Stigmasterol	393±36.2
Campesterol	555±46.0	$\beta$ -Sitosterol	1497±122
Monosaccharide Anhydrides			
Levoglucofan	4473±345		
Polyols			
Glycerol	4155±1743	Arabitol	676±82.4
Erythritol	2699±187	Inositol	1375±103
Alkanes			
Tridecane	70.5±8.26	Tetracosane	121±10.3
Tetradecane	155±8.60	Pentacosane	103±12.1
Pentadecane	156±13.0	Hexacosane	56.4±7.41
Hexadecane	362±30.2	Heptacosane	121±8.14
Heptadecane	493±37.9	Octacosane	26.0±11.1
Octadecane	339±27.7	Nonacosane	276±16.5
Nonadecane	329±26.5	Triacotane	59.7±8.03
Eicosane	197±17.57	Hentriacotane	340±32.8
Heneicosane	215±2.53	Dotriacotane	28.7±6.06
Docosane	163±11.0	Tritriacotane	39.9±5.33
Tricosane	54.4±15.2		
PAHs			
Fluoranthene	8.19±1.64	Benzo(b)fluoranthene	18.8±1.64

**Table 2** (continued)

Target Compounds		Target Compounds	
Pyrene	7.80±1.12	Benzo(k)fluoranthene	16.4±1.61
Benzo(ghi)fluoranthene	3.81±0.52	Benzo(e)pyrene	14.5±1.12
Benz(a)anthracene	1.72±0.29	Benzo(a)pyrene	9.16±0.97
Chrysene/Triphenylene	6.68±0.82	Benzo(ghi)perylene	6.41±1.09
Poly acids			
Malic acid	606±42.67	Glyceric acid	1323±98.3
Aromatic acids			
1,4-Phthalic acid	60.8±6.77	1,2,4-Benzenetricarboxylic acid	6.87±2.75
1,3-Phthalic acid	2.83±1.07	1,3,5-Benzenetricarboxylic acid	4.10±0.80
4-Methoxybenzoic acid	7.57±1.74	1,2,4,5-Benzenetetracarboxylic acid	16.2±2.01
3,4-Dimethoxy benzoic acid	30.7±1.80		

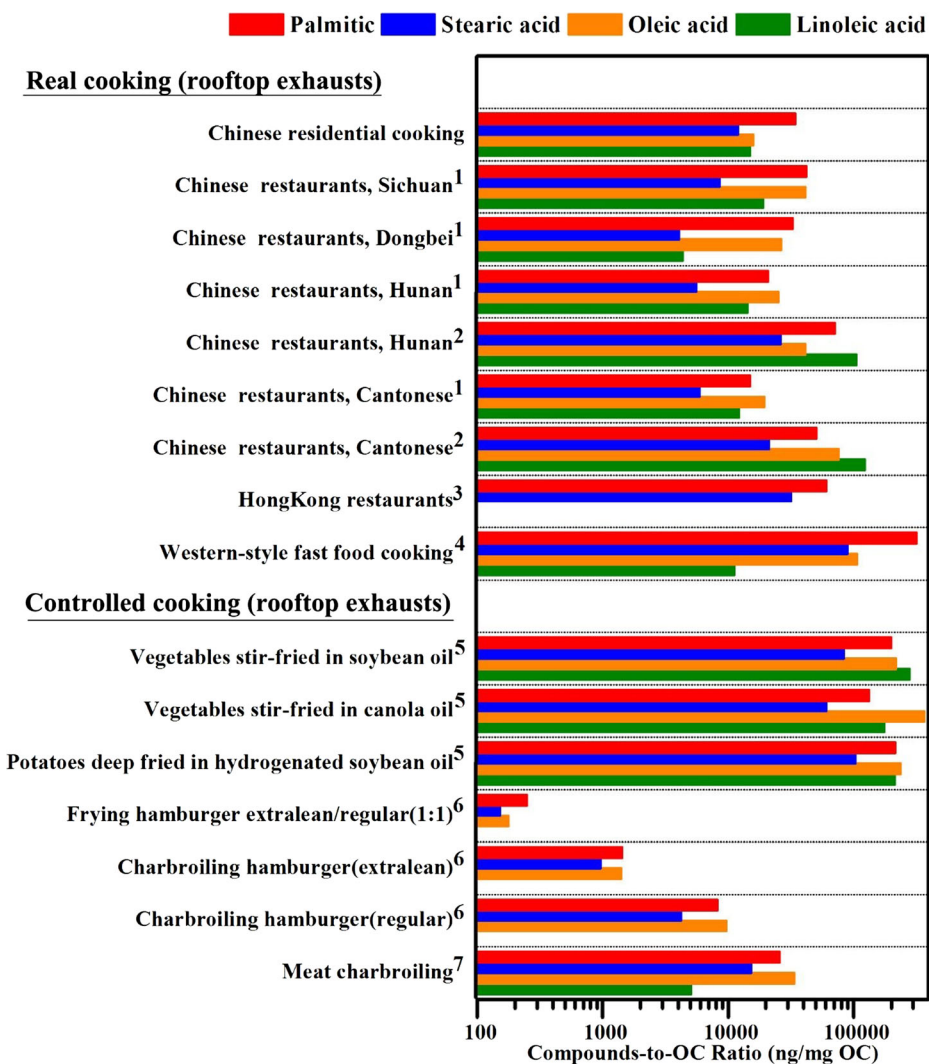
### 3.2.2 Sterols, monosaccharide anhydrides and polyols

Many organics present in ambient aerosols are from biological origin and thus have similar chemical structure with natural chemicals found in living organisms. Such molecules are often termed as molecular biomarkers because of their restricted occurrence, source specificity, molecular stability, and suitable concentration for detection (Mazurek et al. 1987).



**Fig. 3** Pie chart showing the relative abundances of organic homologues detected in the Chinese residential cooking emissions

Sterols are widely present in animal and vegetable tissues. For instance, plant lipid membranes and waxes generally contain C<sub>28</sub> and C<sub>29</sub> phytosterol compounds like  $\beta$ -sitosterol, campesterol and stigmasterol (Oros and Simoneit 2001a, b), while cholesterol is biosynthesized by higher animals and found in all tissues, especially in fats and oils (Rogge et al. 1991). In this study, four kinds of sterols were detected in the residential cooking emissions, among which  $\beta$ -sitosterol was the most abundant, followed by cholesterol, campesterol and stigmasterol (Fig. 5). However, only cholesterol was detectable in meat cooking emissions (Rogge et al. 1991; Schauer et al. 1999; McDonald et al. 2003) and western-style fast food cooking emissions (Zhao et al. 2007b). The abundance of phytosterols from Chinese style cooking was probably related to high temperature cooking (like frying) of



**Fig. 4** Comparison of the concentrations of the four common fatty acids emitted from different cooking studies. <sup>1</sup>Zhao et al. (2007a); <sup>2</sup>He et al. (2004); <sup>3</sup>Hu et al. (2010); <sup>4</sup>Zhao et al. (2007b); <sup>5</sup>Schauer et al. (2002); <sup>6</sup>Rogge et al. (1991); <sup>7</sup>Schauer et al. (1999)

vegetable (He et al. 2004; Zhao et al. 2007a). It should be noted that campesterol was only detected in the present study with the concentration was 555 ng/mg OC. Total concentrations of sterols from the residential cooking (3229 ng/mg OC) in the present study as well as those from Chinese restaurant cooking (He et al. 2004; Zhao et al. 2007a) were over 10 times higher than those from western cooking (Rogge et al. 1991; Schauer et al. 1999; McDonald et al. 2003; Zhao et al. 2007b) (Fig. 5).

Levoglucosan, a kind of monosaccharide anhydrides produced in large quantities during pyrolysis of cellulose, has been applied as an ideal tracer for biomass burning (Simoneit et al. 1999; Simoneit 2002). There were no reports about levoglucosan emitted from meat cooking and seed oil cooking (Rogge et al. 1991; Schauer et al. 1999, 2002), however, this tracer was detected in most Chinese cooking emissions (He et al. 2004; Zhao et al. 2007a, b; Hu et al. 2010), levoglucosan might be produced during high temperature processing of vegetables. The levels of levoglucosan in this study (4473 ng/mg OC in average) were similar to those from Hong Kong restaurants cooking (Hu et al. 2010), but much higher than those from other Chinese restaurant cooking (61.9–776 ng/mg OC) (He et al. 2004; Zhao et al. 2007a, b) (Fig. 5).

Polyols including glycerol, erythritol, arabitol and inositol, were only detected in the residential cooking emissions. The sum of the four species of polyols accounted for 7.66 % of the quantified organic mass, with an average concentration of 8904 ng/mg OC. Glycerol, which is produced by the hydrolysis of triglycerides (Zhu 1995), was the most abundant polyols, with an average concentration of 4155 ng/mg OC.

### 3.2.3 Dicarboxylic acids

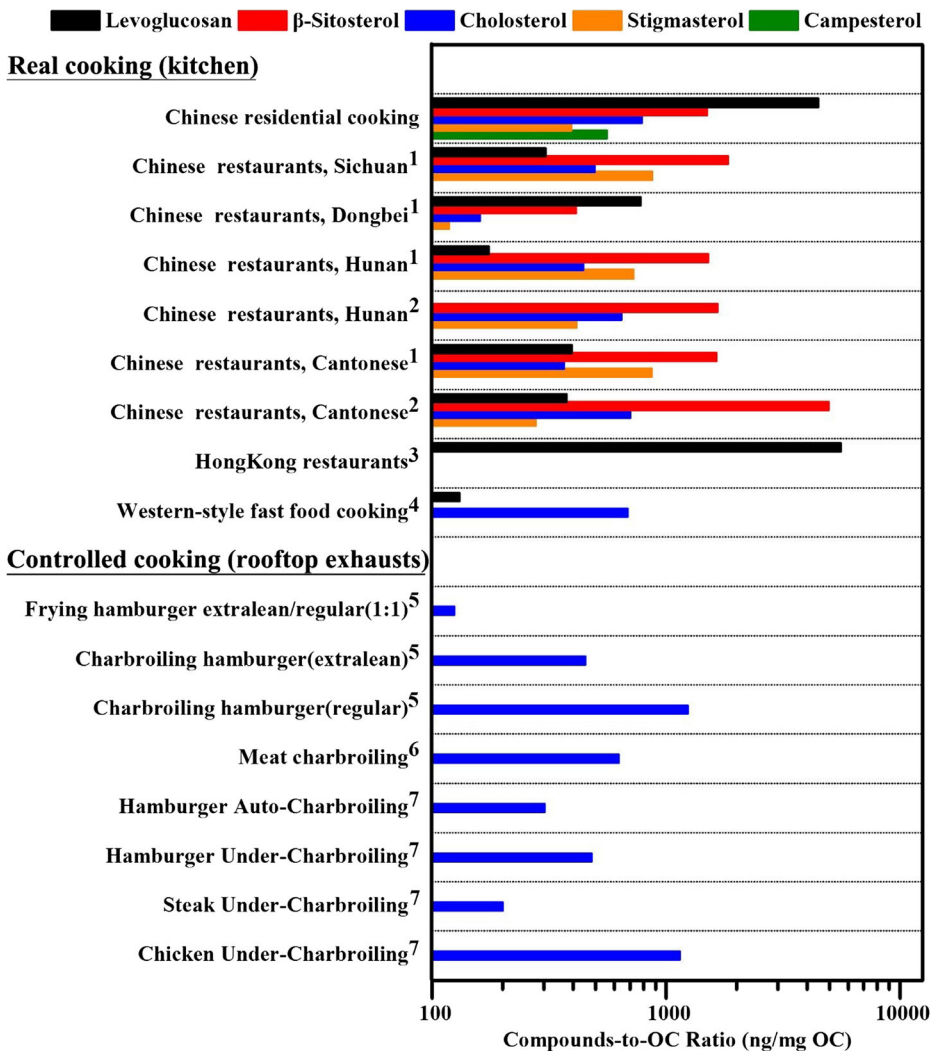
C<sub>6</sub>-C<sub>10</sub> dicarboxylic acids were detected with C<sub>9</sub> as the predominant species (Table 1). The sum of detected dicarboxylic acids accounted for 4.97 % of quantified organic compounds (Fig. 3), which was similar to those reported in cooking emissions from Chinese restaurants (2.07–9.66 %) (He et al. 2004; Zhao et al. 2007a, b). It indicated that although these dicarboxylic acids were normally considered to be secondarily formed through gas-particle transformation in the atmosphere (Rogge et al. 1991), primary emissions like cooking emissions were also these sources. The compositions of dicarboxylic acids in this study resembled those in Chinese restaurant cooking emissions (He et al. 2004; Zhao et al. 2007a), but were quite different from those in western cooking emissions, such as meat cooking and seed oils cooking emissions. Meat cooking produces dicarboxylic acids ranging from C<sub>4</sub> to C<sub>8</sub> (Rogge et al. 1991; Schauer et al. 1999), and seed oils cooking only produces C<sub>6</sub> and C<sub>8</sub> (Schauer et al. 2002).

### 3.2.4 PAHs

The Polycyclic aromatic hydrocarbons (PAHs) are produced from incomplete combustion or pyrolysis of organic substances. PAHs are stable in the environment, and some of species are known as carcinogens (USEPA 1999; IARC 2010).

Only 10 species of PAHs were detected and they together shared a very small fraction (0.08 %) in the quantified compounds in the residential cooking emissions (Table 2). In other cooking emissions studies, PAHs also had little contribution, no more than 2.07 % of quantified compounds as far as we know (Rogge et al. 1991; Schauer et al. 1999, 2002; He et al. 2004; Zhao et al. 2007a, b; Hu et al. 2010).

Like restaurant cooking in Hong Kong (Hu et al. 2010), residential cooking in Guangzhou emitted a little more 5-ring PAHs than other PAHs. However, pyrene was the highest in other



**Fig. 5** Comparison of the concentrations of the major molecular biomarkers emitted from different cooking studies. <sup>1</sup>Zhao et al. (2007a); <sup>2</sup>He et al. (2004); <sup>3</sup>Hu et al. (2010); <sup>4</sup>Zhao et al. (2007b); <sup>5</sup>Rogge et al. (1991); <sup>6</sup>Schauer et al. (1999); <sup>7</sup>McDonald et al. (2003)

Chinese restaurant cooking emissions (He et al. 2004; Zhao et al. 2007a), chrysene was the highest in meat cooking and seed oils cooking emissions (Rogge et al. 1991; Schauer et al. 1999, 2002), and phenanthrene was the highest in chicken and beef charbroiling and grilling (McDonald et al. 2003). These trends were attributed to the types of oil used and the temperatures reached during cooking.

As discussed above, cooking styles could affect emission factors of some organic compounds. Deep frying and charbroiling were found to produce more pollutants, due to high temperature and large amount of oil used during cooking (above 1000 ng/mg OC) (Schauer et al. 2002; McDonald et al. 2003). Cantonese and Sichuan restaurants were found to generate more PAHs (Zhao et al. 2007a). Nonetheless, other styles of cooking including Chinese

residential cooking contributed PAHs with no more than 500 ng/mg OC (Rogge et al. 1991; He et al. 2004; Zhao et al. 2007a, b; Hu et al. 2010).

According to potency equivalency factors (PEF) approach, the cancer risk assessment approach proposed by Yassaa et al. (2001), benzo(a)pyrene (BaP) equivalence factors (BaP<sub>eq</sub>) (carcinogenic PAHs normalized to BaP) in this study were 11.7 ng/mg OC, similar to the results from other styles of restaurant cooking (Rogge et al. 1991; Schauer et al. 1999, 2002; He et al. 2004; Zhao et al. 2007a, b; Hu et al. 2010). However, charbroiling was found to produce much higher levels of BaP<sub>eq</sub> than other the above-mentioned cooking style, which was up to 180 ng/mg OC (McDonald et al. 2003).

### 3.2.5 *n*-Alkanes

C<sub>13</sub>-C<sub>33</sub> normal alkanes were also quantified (Table 2). They contributed only a minor fraction (3.19 %) in the total quantified organic mass (Fig. 3). This fraction was within the range of 0.32–5.97 % reported in other cooking emission studies (Rogge et al. 1991; Schauer et al. 1999, 2002; He et al. 2004; Zhao et al. 2007a, b; Hu et al. 2010).

Higher alkanes (carbon number over 23) showed a distinct odd-to-even carbon preference with an average carbon preference index (CPI) of 3.02 and a maximum level at C<sub>31</sub>, indicating their higher plant wax origin (Simoneit 1986; Rogge et al. 1993). Higher values of CPI were found in previous vegetables cooking studies (9.44–13.8) (Schauer et al. 2002), and lower values (1.08–1.62) were observed in emissions from cooking with little vegetables such as western style fast and meat cooking (Rogge et al. 1991; Zhao et al. 2007b), further indicating the origin of alkanes from vegetables. These high-molecular weight organic compounds volatilized from leaves under higher temperatures during cooking, and then condensed onto existing particles (Zhao et al. 2007a).

### 3.2.6 *Poly acids and aromatic acid*

Poly acids and aromatic acids are considered as secondary oxidation products of organic precursors (Fu et al. 2011). In this study, they were also observed in cooking emissions. Poly acids including glyceric and malic acids accounted for 1.66 % of the total mass of quantified compounds in the residential cooking emissions, while aromatic acids only accounted for 0.11 % of the quantified mass (Fig. 3).

## 3.3 The contribution of cooking emission to ambient OC estimated by tracers

Cholesterol has been widely used as a marker for meat cooking emissions in the CMB model to evaluate the contribution of cooking emissions to ambient organic aerosols (Cass 1998; Robinson et al. 2006; Zheng et al. 2006). Because most people cook their meals at home, the residential emission profiles are more representative than restaurant emission profiles. Because particles from cooking is concentrated in fine and even submicrometre particles (Buonanno et al., 2009; Abdullahi et al. 2013), we assume that the profile in PM<sub>2.5</sub> from cooking is similar to that in TSP. Therefore, with the composition profiles obtained from this study, we applied the tracer-based approach to explore the contribution of cooking emissions to ambient OC in ambient PM<sub>2.5</sub> samples collected at urban site (GZ and GD), suburban sites (PY and HD) and rural sites (KD and WQS) in Guangzhou as mentioned in our previous paper (Zhao et al. 2014).

The percentages of ambient OC contributed by cooking emission can be simply calculated as:

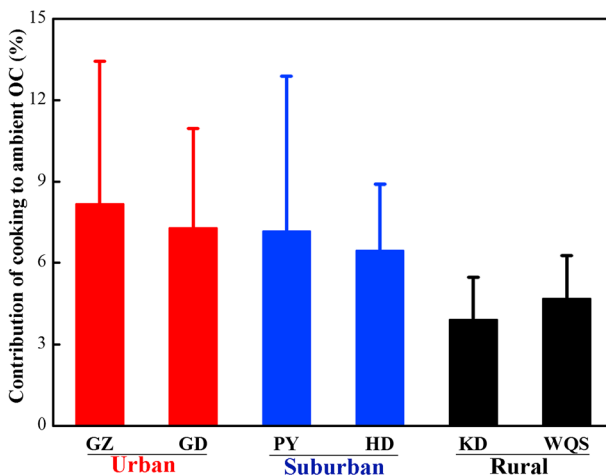
$$\text{OC}\% = \frac{\text{OC}_P \times C_A}{C_P \times \text{OC}_A} \times 100$$

Where  $C_A$  and  $C_P$  are the concentrations of cholesterol in ambient aerosols and cooking emissions, respectively;  $\text{OC}_A$  and  $\text{OC}_P$  are organic carbon contents in ambient aerosols and from residential cooking emissions, respectively.

With the equation, the average percentages of ambient OC contributed by residential cooking emissions were presented in Fig. 6. The urban sites GZ and GD showed relative high cooking emission contributions, with the average of 8.18 % and 7.28 %, respectively; the suburban sites PY and HD showed moderate contributions, with the average were 7.16 % and 6.45 %, respectively; while only 3.90 % and 4.67 % of OC was contributed by cooking emissions at the rural sites KD and WQS, respectively. The result suggested that cooking is an important source of ambient organic aerosols. These estimates in urban sites were comparable to the results from CMB model performed in Guangzhou (Zheng et al. 2011), Hong Kong (Hu et al. 2010) and Shanghai (Feng et al. 2006), but 3 times smaller than those in Beijing (Wang et al. 2009). In addition, the contributions showed obviously spatial trend (urban>suburban>rural), which coincided well with the distribution of the population. Similarly, the contribution of cooking to  $\text{PM}_{2.5}$  mass was also estimated using mass ratio of cholesterol in the cooking emitted aerosols. The results showed that the average contribution of cooking to ambient  $\text{PM}_{2.5}$  was 1.69 %. It is noted however, because of the different particle size of ambient samples and source samples, the accuracy of this estimation is limited.

#### 4 Conclusion

TSP samples from residential cooking emissions in an urban site of Guangzhou, south China were collected and the organic compositions of these samples were analyzed. The average



**Fig. 6** Contribution of residential cooking to ambient OC at six sites

mass concentration of TSP emitted from the Chinese residential cooking monitored in rooftop exhausts was  $138 \mu\text{g}/\text{m}^3$ . This result was lower than those monitored in residential kitchen and in rooftop exhausts of Chinese restaurant. OM, as the major component, accounted for 66.9 % of TSP mass in Chinese residential cooking emissions.

Over 90 organic species were identified and quantified, accounting for 14.5 % of OC altogether. The levels and chemical compositions of the organic compounds from the Chinese residential cooking showed small differences with those from Chinese restaurant cooking, however, obvious differences with those from western-style cooking. Fatty acids, including saturated acids and unsaturated acids, were the dominant species in Chinese residential cooking, and constituted 75.7 % of the total mass of quantified compounds, with palmitic acid as the predominant species. Sterols, monosaccharide anhydrides and polyols were also significant compositions and they shared 14.3 % in the quantified organic compounds. Phytosterols were only detected from Chinese-style cooking, and campesterol and polyols were only detected in the present study. Dicarboxylic acids accounted for 4.97 % of quantified organic compounds, with  $\text{C}_9$  being the predominant one. Whereas PAHs and alkanes contributed only small fractions to the quantified compounds.

The contributions of cooking emissions to ambient OC in urban, suburban and rural sites, roughly calculated by typical cooking tracer cholesterol, ranged from 3.90 % in rural area to 8.18 % in urban site. The spatial distribution of cooking contributions was in accord with the distribution of population.

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