Emission of fine organic aerosol from traditional charcoal broiling in China

Ximei Hou • Guoshun Zhuang • Yanfen Lin • Juan Li • Yilun Jiang • Joshua S. Fu

Received: 12 September 2007 / Accepted: 8 July 2009 /

Published online: 17 July 2009

© Springer Science + Business Media B.V. 2009

Abstract The indoor PM_{2.5} aerosol samples for charcoal broiling source under Chinese traditional charbroiling and the ambient fine aerosols samples (PM_{2.5}) were collected in Beijing to investigate the characteristics of the charcoal broiling source and its impact on the fine organic aerosols in the atmosphere. The concentrations of 20 species of the trace organic compounds, including polycyclic aromatic hydrocarbons (PAHs), fatty acids, levoglucosan, and cholesterol in PM_{2.5} were identified and quantified by GC/MS. The total PAHs and fatty acids emitted from charcoal broiling to PM_{2.5} were 8.97 and 87,000 ng mg⁻¹ respectively. The concentrations of the light molecular weight (LMW) 3and 4-ring PAHs were much higher than those of the high molecular weight (HMW) 5- and 6-ring PAHs. Fatty acids were the most abundant species in source profile, accounting for over 90% of all identified organic compounds. More polyunsaturated fatty acid (linoleic acids) than the saturated fatty acid (stearic acids) emitted in the cooking. Charcoal broiling is a minor source of PAHs compared to the source of biomass burning. Comparing the ratios of levoglucosan/fatty acid and levoglucosan/cholesterol in the charcoal broiling samples to the ambient samples, it is evident that meat cooking is an important source of fatty acids, but a less important source of cholesterol. Cooking, as one of the source of fine organic particles, plus other anthropogenic sources would be related to the formation of the severe haze occurred and spread over the urban atmosphere in most of the cities of China in the past several years.

X. Hou · G. Zhuang () · Y. Lin · J. Li · Y. Jiang

Center for Atmospheric Chemistry Study, Department of Environmental Science & Engineering,

Fudan University, Shanghai 200433, China

e-mail: gzhuang@fudan.edu.cn

X. Hou · G. Zhuang

Center for Atmospheric Environmental Study, Department of Chemistry, Beijing Normal University, Beijing, 100875, China

J. S. Fu

Department of Civil and Environmental Engineering, The University of Tennessee, Knoxville, TN 37996, USA



Keywords Charcoal broiling · Organic aerosol · PAH · Fatty acids · Levoglucosan

1 Introduction

For the past decades, the haze has occurred and spread over the Central and Eastern China (Kaiser and Qian 2002) due to increasing vehicle pollution, coal combustion, domestic heating and cooking. The severe haze not only damages health and agriculture, but also changes the earth's radiation balance. Haze is mainly resulted from anthropogenic emissions of particles or gases transformed to particles after emission, of which the organic aerosols are one of the important components. It's reported that the carbonaceous material contributed about 30% to aerosol optical thickness (a measure of aerosol column concentration) in the urban haze of the eastern United States (Novakov et al. 1997; Kaufman et al. 2002). As a main emission source of the organic pollutants in urban atmosphere, meat cooking emits fine particles, of which organic aerosol account for 68~73% (Hildemann et al. 1991).

In the previous studies related to the emission from cooking, the detailed organic chemical composition of fine particles from frying, natural gas charbroiling, and Chinese traditional cooking in the kitchen has been examined (Rogge et al. 1991; Schauer et al. 1999a; He et al. 2004). However, the study on Chinese charcoal broiling, which has recently been very common and popular, even everywhere on the sides of the streets in most cities, for the daily food in China, has not been addressed. The barbecue stalls are common on the street sides in China, while many charcoal broiling activities are made indoor with simple exhaust devices and also often are conducted outdoors by restaurants and families. Although the exhaust hoods are used to exhale the smoke on most charcoal broiling grills, both indoor air and outdoor ambient atmosphere are severely polluted. Thus, it is very necessary to study on the organic compound emission from charcoal broiling. The comprehensive information on the chemical composition of organic aerosols can help to estimate the contribution of charbroiling source to the atmospheric fine aerosols, and to understand further its impact on the air quality.

In this study, we carried out the source sampling in accordance with the Chinese traditional charcoal broiling, analyzed PAHs, fatty acids, levoglucosan, and cholesterol, which were also identified in urban atmospheric aerosols samples, and probed the chemical composition, the characterization of source profile, and the influences of charcoal broiling on the ambient atmosphere.

2 Experimental

2.1 Source testing procedure

The entire source sampling experiment was conducted in a clean room (7 m²), which strictly followed the procedures of the traditional charbroiling cooking. Before sampling, the indoor aerosols were sampled as blank background. The regular mutton with several kinds of condiments, salt, capsicum spice, and cuminum powder, which are the main condiments of charbroiling mutton, was charbroiled in a common commercial-scale charbroiling grill with 0.5 kg of charcoal in this experiment. The sliced mutton was bunched on the bamboo brochettes. The meat was cooked on the grill to be "well-done". During the process of charbroiling, the condiments were spilled on the meat several times as traditional Chinese broiling does.



Three PM_{2.5} samplers (Beijing Geological Instrument-Dickel Co., Ltd.) were located on the floor and 1 m away from the grill. As broiling started, the samplers were operated at flow rate 77.59 L min⁻¹. The fine particle was collected onto the quartz filter holders, which composed of two quartz filters: the front filter was used to collect the fine particle and the backup filters was used to collect the semi-volatile organic compounds, which were vaporized from the front filter. The quartz filters (90 mm, Whatman Company, UK) had been baked out at 500°C for 2 h prior to sampling to minimize the background of the organic contaminants. After completion of the sampling, the sample filters were put in the polyethylene plastic bags and stored at -20°C in a refrigerator to avoid possible bacterial degradation.

2.2 Extraction

The samples were ultrasonically extracted thrice with 20 ml of 1:1 dichloromethane/acetone (HPLC grade, Fisher, USA) each for 15 min, and then the extracts were combined together and filtered with glass fiber filter to remove those insoluble particles. The filtrates were concentrated to 3 ml by a rotary evaporator under gentle vacuum. These concentrated filtrates were divided into three fractions and spiked with deuterated tetracosane (n-C₂₄D₅₀, Alltech, USA) and hexamethyl benzene (Aldrich Chem. Co., USA) for the internal standards and recovery tests of fatty acids, levoglucosan, cholesterol, and PAHs. All three fractions were reduced to near dryness by gentle evaporation with a steam of high purity N₂, and re-dissolved in hexane. The fraction for measuring PAHs was cleaned up on silica SPE cartridges (500 mg, 6 ml) (Phenomenex, Torrance, CA, USA), then the final concentrated filtrates were adjusted to 500 µl by adding hexane for GC/MS analysis. The fraction for measuring fatty acids was derivatized by 500 µl of 10% BF3-methanol (Supelco, USA) to obtain the corresponding methyl esters for 30 min at 80°C. The uncertainty of derivatization of fatty acids and other trace organic compounds was less than 10% (Hou et al. 2006). The fraction for measuring levoglucosan and cholesterol was derivatized by bis-(trimethylsilyl)trifluoroacetamide (BSTFA, Supelco, USA) to convert to their trimethylsilyl derivatives for 3 h at 70°C. After extraction and dryness, the derivatives were re-dissolved in hexane and concentrated to 500 μl until the analysis by GC/MS.

2.3 GC/MS analysis

12 PAHs, seven fatty acids and two tracers (levoglucosan and cholesterol) in samples were identified and quantified by using a Trace GC//MS spectrometer (ThermoQuest, Finnigan San Jose, CA, USA) with a DB-5MS capillary column (J&W Scientific, Folsom, CA, USA, 30 m×0.25 mm I.D., 0.25 um film thickness). High purity helium (99.999%) was used as carrier gas at a flow rate of 1.0 ml min⁻¹. The quantification of PAHs was conducted with selective ion monitoring (SIM) mode and its GC temperature program was from 100°C (2 min) to 200°C at 10°C min⁻¹ and hold at 200°C for 3 min, then ramp to 290°C at 5°C min⁻¹ and hold at 290°C for 7 min. The temperature program for fatty acids analysis was first hold at 60°C for 2 min, ramp to 60°C at 20°C min⁻¹ and hold at 160°C for 5 min, then ramp to 250°C at 10°C min⁻¹ and hold at 250°C for another 3 min. The temperature program for cholesterol and levoglucosan analysis was first hold at 65°C for 2 min, ramp to 120°C at 30°C min⁻¹, then ramp to 300°C at 10°C min⁻¹ and hold at 300°C for 10 min.

The target compounds were identified by comparison with the retention times and the mass fragmentation patterns of those authentic standards, under the help of the NIST98 (National Institute of Standards and Technology) standard library. The Relative response



factors were calculated from the GC/MS analysis of the standard compounds. The quantification and identification standards were as below: 1. 12 PAHs (Chem. Service), Fluorine (99%, Flu), Anthracene (98.1%, Anth), Fluoranthene (98.0%, Flua), Pyrene (98%, Pyr), Benzo[a]anthracene (98%, B[a]A), Chrysene (98%, Chr), Benzo[b]fluoranthene (99%, B[b]F), Benzo[k]fluoranthene (97.5%, B[k]F), Benzo[a]pyrene (99.2%, B[a]P), Dibenz[a,h]anthracene (98.3%, Dib[a,h]A), Benzo[ghi]perylene (99.2%, B[ghi]P), and Indeno[1,2,3-cd]pyrene (99.5%, IndP); 2. seven methylated fatty acids (Sigma), lauric acid, myristic acid, hexadecanoic acid (palmitic acid), heptadecanoic acid, linoleic acid, oleic acid, and octadecanoic acid (stearic acid); 3. levoglucosan (Aldrich) and cholesterol (Sigma). The detection limits of these organic compounds were 0.6~500 pg. The quality assurance protocol was as follows: The procedural blanks and field blanks taken from the source samples during operation were examined for contaminants by the same experimental procedures (Mazurek et al. 1987). The target compounds of procedural and field blank were not found or below the detection limit. Recovery tests were carried out through the same experimental procedure, i.e. by spiking the blank filters with a standard mixture containing the 12 PAHs, 4 Fatty Acids (hexadecanoic acid, linoleic acid, oleic acid and octadecanoic acid, Sigma), and levoglucoasan, cholesterol. The recovery of each target compound was in the range of $80\sim96\%$, and the accuracy for the quantification was within $\pm20\%$.

2.4 Urban fine particle sampling

The atmospheric fine aerosols samples (PM_{2.5}) were collected daily for 12 h at Beijing Normal University (sampling height: 30 m) during two periods (July 18~August 13, 2004; December 15, 2004~January 19, 2005), using medium volume samplers, at a flow rate of 77.59 L min⁻¹ on the quartz filters. Three sequential samples were combined to one to detect all of those trace organic compounds interested in this study, as the concentration of a few trace organic compounds in each sample filter was too low to be detected. The analysis protocol of ambient samples was the same as that used in charbroiling source profile study as mentioned above.

3 Results and discussion

The typical selected mass fragmentograms for PAHs, total ion current traces for fatty acids, levoglucosan, and cholesterol from GC/MS analyses in charbroiling source were shown in Figs. 1, 2 and 3.

Under experimental conditions, some semi-volatile organic compounds could condense on the preexisting indoor aerosols before filter sampling, because the organic compounds emitted from the charcoal broiling smoke could be mixed and diluted with the indoor air. That is, the source test would be influenced by the indoor aerosols. In our study, two low molecular weight PAH (Fluorethene, Pyrene) and all fatty acids except heptadecanoic acid and linoleic acid were found in the indoor samples, which were at $2\sim4$ orders of magnitude of the concentrations lower than those detected on the front filters. This result indicated that the indoor dilution air was very low in contaminants.

Meanwhile, because of atmospheric dilution and cooling effects, the partitioning between gas-phase and particle-phase of the semi-volatile organics in smoke aerosols could be disturbed during filter sampling (Ligocki and Pankow 1989). Therefore, the backup filter still collected some organic aerosols, which was due to adsorption of gaseous semi-volatile compounds onto the backup filter, or re-adsorption of the particle-phase semi-



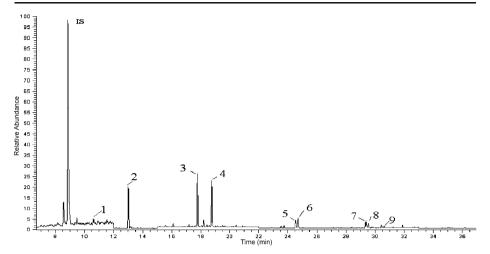


Fig. 1 Selected mass fragmentograms from GC/MS analysis for PAHs. 1, Fluorine; 2, Anthracene; 3, Fluoranthene; 4, Pyrene; 5, Benzo[a]anthracene; 6, Chrysene; 7, Benzo[b]fluoranthene; 8, Benzo[k] fluoranthene; 9, Benzo[a]pyrene; IS, Internal standard (C₆(CH₃)₆)

volatile organics from volatilization on the front filters (Fitz 1990; Hildemann et al. 1991). In order to understand the particulate on the backup filters, the results of front and backup filters were compared (see Fig. 4a–b). Figure 4a showed that only the LMW PAHs presented in the profile of backup filters and the concentrations of 3-ring PAHs were higher than the ones of the front filters, while the HMW PAHs were opposite. Figure 4b presented that the less volatile C18 series fatty acids (linoleic acids, oleic acid, and stearic acid) were at lower concentrations than the more volatile fatty acids of ≤C16 found on the backup samples, whereas the palmitic acid and the C18 series fatty acids were of considerable amounts on the front filters. This suggested that those PAHs less volatile with higher

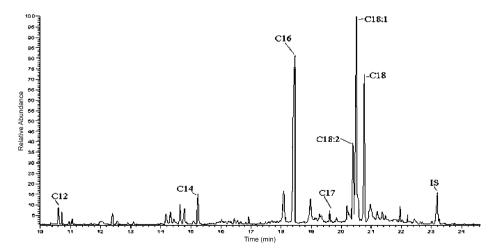


Fig. 2 Total ion GC/MS chromatogram for fatty acids as methyl esters. C12, lauric acid; C14, myristic acid; C16, palmitic acid; C17, heptadecanoic acid; C18:2, linoleic acid; C18:1, oleic acid; C18, stearic acid; IS, Internal standard, (n-C₂₄D₅₀)



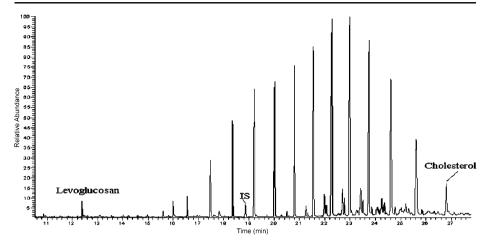


Fig. 3 Total ion GC/MS chromatogram for levoglucosan and cholesterol as trimethylsilyl derivatives. IS, Internal standard, (n-C₂₄D₅₀)

molecular weight and fatty acids were predominant in the particle phase collected on the front filters. The total concentrations of the semi-volatile organics on backup filters were $0.1 \sim 1\%$ of those on the front filters.

3.1 Composition of the smoke aerosol in the charcoal broiling

3.1.1 Polycyclic aromatic hydrocarbons (PAHs)

PAHs are the pollutants from incomplete combustion, which are of special interest due to their toxicity, carcinogenicity, and ubiquitous presence in the environment (Halsall et al. 1994; Schauer et al. 2003). The findings of Panalaks (1976) suggested that the concentrations of PAHs in charcoal-broiled meat were proportional to the fat content of the raw meat (Panalaks 1976). The rendered fat fell into the hot charcoals and was pyrolyzed, forming PAHs, which were then volatilized and partly re-deposited on the meat surface.

In our experiment, the semi-volatile of 3- and 4-ring PAHs and the particle-phase of 5-ring PAHs were detected in the front filters (see Fig. 1). The semi-volatile fluorine and anthracene were predominant in the gaseous phase depending on the temperature and aerosol concentration (Schauer et al. 2003). Moreover, the high molecular weight of 6-ring PAHs were not detected in the charbroiling source samples, which was similar to the earlier study on charcoal broiling source (Dyremark et al. 1995). The following discussion would be focused on the 4-ring and 5-ring PAHs. The concentrations of PAHs and other organic compounds in the emitted fine particles were listed in Table 1. The total concentration of PAHs was 8.97 ng mg⁻¹. The obvious distribution characteristic was that the concentrations of 4-ring PAHs were much higher than those of 5-ring PAHs, which was in agreement with the previous studies (Rogge et al. 1991; Dyremark et al. 1995; Schauer et al. 1999a).

For comparison, the chemical composition of PAHs from this study with the earlier investigations on charcoal grilling and natural gas charbroiling hamburgers source was shown in Fig. 5. The highest emission concentrations among PAHs in this study were Fluoranthene and Pyrene, which was consistent with the charcoal grilling meat source (Dyremark et al. 1995). However, the highest emission rates among PAHs were chrysene/



Fig. 4 Emission profiles of the organic compounds in the charcoal broiling source. a, PAHs; b, fatty acids

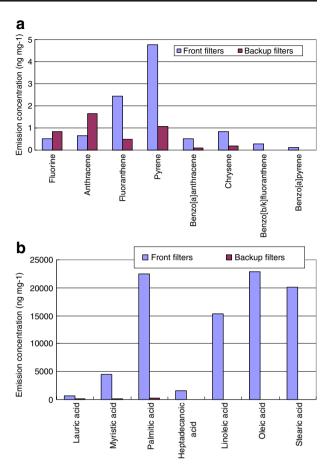
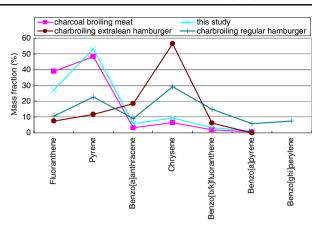


Table 1 Concentrations of organic compounds in fine particles from the charcoal broiling source

PAHs	Conc.(ng mg ⁻¹)	Fatty acids	Conc.(ng mg ⁻¹)	
Fluoranthene	2.45	Lauric acid	609.22	
Pyrene	4.76	Myristic acid	4,500	
Benzo[a]anthracene	0.51	Hexadecanoic acid	22,000	
Chrysene	0.84	Heptadecanoic acid	1,500	
Benzo[b/k]fluoranthene	0.29	Linoleic acid	15,000	
Benzo[a]pyrene	0.13	Oleic acid	230,000	
Indeno[1,2,3-cd]pyrene	0	Octadecanoic acid	20,000	
Dibenz[a,h]anthracene	0	Total fatty acids	87,000	
Benzo[ghi]perylene	0	Levoglucosan	103.61	
Total PAHs	8.97	Cholesterol	58.18	



Fig. 5 Relative contribution of each compound to the sum of PAHs in the charbroiling source. Data of charbroiling extra-lean hamburger and charbroiling regular hamburger from Rogge et al. 1991; data of charcoal broiling meat from Dyremark et al. 1995



triphenylene in natural gas charbroiling regular hamburgers and in extra-lean hamburgers sources (Rogge et al. 1991). The differences in distribution patterns for these charbroiling sources maybe resulted from the different heat sources: charcoal or natural gas. For charcoal combustion, Fluoranthene and Pyrene were of the highest emission rates, while for natural gas combustion chrysene/triphenylene were of (Dyremark et al. 1995; Rogge et al. 1993a), which clearly indicated that the heat source (such as charcoal) had much more impact on the PAHs distribution of meat charbroiling source profile. The different composition characterization, as a result mainly due to the different fuel, indicated that the source characteristics of PAHs from charcoal grilling meat were mainly influenced by the combustion of the charcoal itself (Dyremark et al. 1995). B(ghi)P was not detected in this study, while it did exist in charbroiling regular hamburgers source profiles, which might because of the effect of the various fat contents in raw meat (Rogge et al. 1991). Although this point of view could not be confirmed by this study due to the unknown fat content of the mutton used, the fact that Benzo[ghi]perylene was only presented in charbroiling hamburgers source with a high fat content (20%) would provide supportive evidence for this point.

3.1.2 Fatty acids

The emissions of organic compound from the cooking process were dominated by fatty acids, especially meat cooking. In raw meat, unsaturated and saturated fatty acids exist in the form of phospholipids and triglycerides. When cooking meat, fatty acids were liberated or formed by hydrolysis and thermal oxidation (Rogge et al. 1991; Nolte et al. 1999; He et al. 2004). The total concentration of the quantified fatty acids was 87,000 ng mg⁻¹ in the charbroiling source profile in this study. The concentrations of the emitted oleic acid and palmitic acid were 23,000 ng mg⁻¹ and 22,000 ng mg⁻¹ respectively, the highest among those fatty acids, which was in agreement with the previous studies (Rogge et al. 1991; Schauer et al. 1999a).

Rogge et al. (1991) proposed that the distribution and emission rates of fatty acids in meat cooking source would be markedly influenced by the fat content and composition of fatty acids in raw meat (Rogge et al. 1991; Schauer et al. 1999a). Moreover, after cooking the significant change of the composition of fatty acids was the relative ratios of polyunsaturated fatty acids to the saturated (Candela et al. 1996). The concentration of the saturated stearic acid (C18) and polyunsaturated linoleic acid (C18:2) from this study with



the previous meat cooking source tests and in the raw meat served as cooking materials were listed in Table 2. The ratios of linoleic acid to stearic acid (C18:2/C18) in charcoal broiling mutton (from this study), in gas charbroiling hamburgers (Schauer et al. 1999a), in frying chicken and pork (Candela et al. 1996), and in the corresponding raw meats were shown in Fig. 6. Firstly, the ratios of C18:2/C18 in all cooking profiles were higher than the ones in raw meat and in fat droplets, which indicated that after meat cooking the relative ratio of saturated fatty acid (C18) decreased and polyunsaturated fatty acid (C18:2) increased (Candela et al. 1996). Secondly, increase of ratios in charbroiling source (including charcoal and gas charbroil) was not as distinctly as the one in frying chicken and pork source. Since the liquid grease droplets melt from fat depots due to heating and fell onto the broiling charcoal or into the gas flame, the fatty acids of fat droplets would be vaporized or splattered into the smoke aerosols simply (Rogge et al. 1991), the contribution from the fat droplets in raw meats might be a significant factor for the indistinct change of the ratio (C18:2/C18) in the broiling charcoal.

3.1.3 Levoglucosan and cholesterol

Levoglucosan, the pyrolysis products of cellulous in biomass burning, was detected in the charcoal broiling samples (58.18 ng mg⁻¹). Levoglucosan is commonly used as a tracer for wood combustion in urban atmosphere (Simoneit 1986; Schauer and Cass 2000; Schauer et al. 1996, 2001; Zheng et al. 2002). Levoglucosan wasn't found in frying and charbroiling sources (Rogge et al. 1991; Schauer et al. 1999a), while it was detected in Chinese cooking source profiles with an appreciable amounts in the source emission (He et al. 2004). The cooking process and cooking materials were definitely different between Chinese cooking and American cooking. In China, the vegetable was cooked with the same way as the meat, such as frying, stir-frying. The high temperature during cooking process would lead to the emission of levoglucosan due to thermal degradation of vegetable cellulous. In charcoal broiling source, cumimum cyminum and capsicum spice (powder of dry vegetable) used as condiments were spilled on the mutton (Cremer and Eichner 2000), and the condiments fell onto the broiling charcoal and decomposed to form levoglucosan.

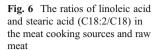
Cholesterol, belonging to the group of steroids, is biosynthesized by higher-class of animals. It is found in all body tissues, especially in animal fats and oils (Rogge et al. 1991; Nolte et al. 1999), which was detected as high as 103.61 ng mg⁻¹ in this study. Cholesterol could be used as an useful molecular marker for cooking operations (meat cooking or cooking of other cholesterol-containing foods) (Rogge et al. 1991; Schauer et al. 1999a; Nolte et al. 1999).

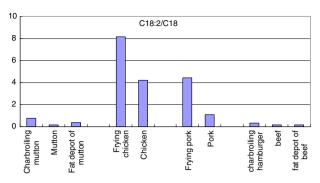
Table 2 Concentrations of linoleic acid and stearic acid from the meat cooking and in raw meat

Ref.	Charcoal broiling mutton ^a	Mutton ^b	Fat depot of mutton ^b	Frying chicken ^c	Chicken ^c	Frying pork ^c	Pork ^c	Charbroiling hamburger ^d	Beef ^e	Fat depot of beef ^b
Unit	ng/mg	g/100 g	g/100 g	g/100 g	g/100 g	g/100 g	g/100 g	mg/kg	mg/g	g/100 g
Linoleic acid (C18:2) Stearic acid (C18)	15,000 20,000	3.28 18.6	4.2 11.55	4.57 0.56	0.46 0.11	7.57 1.71	1.1 1	32,000 97,000	89 507	2.37 14

^a This study; ^b Banskalieva et al. (2000); ^c Candela et al. (1996); ^d Rogge et al. (1991); ^e Wood and Enser 1997).







3.2 Comparison of the source profile with ambient aerosol

The average concentrations of organic compounds identified in the ambient samples of Beijing were shown in Table 3. Figure 7 illustrated the relative contributions of each compound to the overall mass concentration of PAHs in ambient sample and source profile. The highest contributions were Fluoranthene and Pyrene in charcoal broiling source profile, while Benzo[b,k]fluoranthene in ambient aerosols. The LMW 4-ring PAHs dominated in source samples, while the HMW 5- and 6-ring PAHs dominated in ambient aerosols. The distinct difference of PAHs distribution between the source profile and atmospheric aerosols indicated that charbroiling source was not the main emission source of PAHs in atmospheric aerosols. The relative depletion of the LMW PAHs in atmospheric aerosols could be due to enhanced photodecomposition or other atmospheric chemical degradation (Rogge et al. 1991; Schauer et al. 2003). The enrichment of the HMW PAHs in ambient aerosols suggested that other anthropogenic combustion sources of PAHs, such as vehicle exhaust and coal combustion, would be the important contributions in the ambient aerosols (Rogge et al. 1993b, 1997; Schauer et al. 2003).

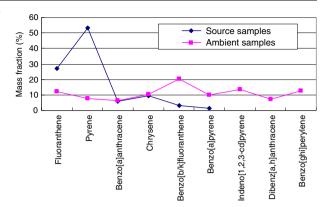
In Fig. 8, the distribution of fatty acids in ambient samples was substantially in agreement with that in source samples, though the relative ratios of C18:2 and C18:1 in ambient samples were lower than those in the source profile. This result indicated the important contribution of the charcoal broiling emission to atmospheric fatty acids. Certainly, the fatty acids in ambient atmosphere, particularly palmitic acid and stearic acid,

Table 3 The average concentrations of the organic compounds in PM_{2.5} in Beijing

PAHs	Conc.(ng m ⁻³)	Fatty acids	Conc.(ng m ⁻³)		
Fluoranthene	7.19	Lauric acid	1.74		
Pyrene	4.43	Myristic acid	6.51		
Benzo[a]anthracene	3.73	Hexadecanoic acid	21.28		
Chrysene	6.12	Heptadecanoic acid	1.76		
Benzo[b/k]fluoranthene	12.14	Linoleic acid	6.22		
Benzo[a]pyrene	5.83	Oleic acid	19.14		
Indeno[1,2,3-cd]pyrene	8.04	Octadecanoic acid	19.5		
Dibenz[a,h]anthracene	4.18	Total fatty acids	76.16		
Benzo[ghi]perylene	7.61	Levoglucosan	18.22		
Total PAHs	59.29	Cholesterol	2.24		



Fig. 7 Relative contribution of each compound to the sum of PAHs in the source samples and ambient samples



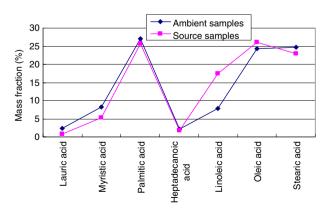
are also emitted from vehicle exhaust (Rogge et al. 1993b; Schauer et al. 1999b), road dust (Rogge et al. 1993c), cigarette smoke and fuel oil combustion (Rogge et al. 1994, 1997). The unsaturated fatty acids (C18:2 and C18:1) in the atmosphere were depleted possibly because of the reaction with the ambient ozone (Nolte et al. 1999).

With the comparison of Table 1 and Table 3, it could be seen that charcoal broiling is a minor source of PAHs compared to the source of biomass burning. Also, by comparing the ratios of levoglucosan/fatty acid and levoglucosan/cholesterol in the charcoal broiling samples to the ambient samples, it is evident that meat cooking is an important source of fatty acids, but a less important source of cholesterol. In addition, the sum of the total seven fatty acids was as high as 76.16 ng m⁻³, which could account for 1–2% of the total OC in PM_{2.5} in Beijing (Dan et al. 2004). As mentioned above, fatty acids were mostly from cooking, thus it was evident that cooking is one of the major sources of the organic aerosol in the fine particles.

4 Conclusions

The total PAHs and fatty acids emitted from charcoal broiling to PM_{2.5} were 8.97 and 87,000 ng mg-1 respectively. The concentrations of the light molecular weight (LMW) 3-and 4-ring PAHs were much higher than those of the high molecular weight (HMW) 5- and

Fig. 8 Relative contribution of each compound to the sum of fatty acids in the source samples and ambient samples





6-ring PAHs. Fatty acids were the most abundant species in source profile, accounting for over 90% of all organic compounds identified. More polyunsaturated fatty acid (linoleic acids) than the saturated fatty acid (stearic acids) emitted in the cooking. Charcoal broiling is a minor source of PAHs compared to the source of biomass burning. Comparing the ratios of levoglucosan/fatty acid and levoglucosan/cholesterol in the charcoal broiling samples to the ambient samples, it is evident that meat cooking is an important source of fatty acids, but a less important source of cholesterol. Cooking, as one of the source of fine organic particles, plus other anthropogenic sources would be related to the formation of the severe haze occurred and spread over the urban atmosphere in most of the cities of China in the past several years.

Acknowledgements This work was supported by the National Key Project of Basic Research of China (Grant No.2006CB403704), Beijing Natural Science Foundation (Grant No. 8041003), and National Natural Science Foundation of China (Grant Nos. 20877020, 40575062, and 40599420).

References

- Banskalieva, V., Sahlu, T., Goetsch, A.L.: Fatty acid composition of goat muscles and fat depots: a review. Small Rumin. Res. 37, 255–268 (2000)
- Candela, M., Astlasaran, I., Bello, J.: Effect of frying on the fatty acid profile of some meat dishes. J. Food Compos. Anal. 9, 277–282 (1996)
- Cremer, D.R., Eichner, K.: Formation of volatile compounds during heating of spice Paprika (Capsicum annuum) Powder. J. Agric. Food Chem. 48, 2454–2460 (2000)
- Dan, M., Zhuang, G., Li, X., Tao, H., Zhuang, Y.: The characteristics of carbonaceous species and their sources in PM2.5 in Beijing. Atmos. Environ. 38, 3443–3452 (2004)
- Dyremark, A., Westerholm, R., Övervik, E., Gustavsson, J.: Polycyclic Aromatic hydrocarbon (PAH) emissions from charcoal grilling. Atmos. Environ. 29, 1553–1558 (1995)
- Fitz, D.R.: Reduction of the positive organic artifact on quartz filters. Aerosol Sci. Tech. 12, 142–148 (1990) Halsall, C.J., Coleman, P.J., Davis, B.J., Burnett, V., Waterhouse, K.S., Harding-Jones, P., Jones, K.C.: Polycyclic aromatic hydrocarbons in U.K. urban air. Environ. Sci. Technology 28, 2380–2386 (1994)
- He, L., Hu, M., Huang, X., Yu, B., Zhang, Y., Liu, D.: Measurement of emissions of fine particulate organic matter from Chinese cooking. Atmos. Environ. 38, 6557–6564 (2004)
- Hildemann, L.M., Markowski, G.R., Cass, G.R.: Chemical composition of emissions from urban sources of fine organic aerosols. Environ. Sci. Technology 25, 744–759 (1991)
- Hou, X., Zhuang, G., Sun, Y., An, Z.: Characteristics and sources of polycyclic aromatic hydrocarbons and fatty acids in PM_{2.5} aerosols in dust season in China. Atmos. Environ. **40**, 3251–3262 (2006)
- Kaiser, D.P., Qian, Y.: Decreasing trends in sunshine duration over China for 1954–1998: indication of increased haze pollution? Geophys. Res. Lett. 29, 2042–2045 (2002)
- Kaufman, Y.J., Tanré, D., Boucher, O.: A satellite view of aerosols in the climate system. Nature 419(215), 223 (2002)
- Ligocki, M.P., Pankow, J.F.: Measurements of the gas/particle distributions of atmospheric organic compounds. Environ. Sci. Technology 23, 75–83 (1989)
- Mazurek, M.A., Simoneit, B.R.T., Cass, G.R., Gray, H.A.: Quantitative high-resolution gas chromatography and high-resolution gas chromatography / mass spectrometry analysis of carbonaceous fine aerosol particles. Int. J. Envir. Analyt. Chem. 29, 119–139 (1987)
- Nolte, C.G., Schauer, J.J., Cass, G.R., Simoneit, B.T.: Highly polar organic compounds present in meat smoke. Environ. Sci. Technology. 33, 3313–3316 (1999)
- Novakov, T., Hegg, D.A., Hobbs, P.V.: Airborne measurements of carbonaceous aerosols on the East Coast of the United States. J. Geophys. Res. 102, 30023–30030 (1997)
- Panalaks, T.: Determination and identification of polycyclic aromatic hydrocarbons in smoked and charcoal-broiled food products by high pressure liquid chromatopphy. J Environ. Sci. Health B. 11(4), 299–315 (1976)



- Rogge, W.F., Hildemann, L.M., Mazurek, M.A., Cass, G.R., Simoneit, B.R.T.: Sources of fine organic aerosol: 1. Charbroilers and meat cooking operations. Environ. Sci. Technology 25, 1112–1125 (1991)
- Rogge, W.F., Hildemann, L.M., Mazurek, M.A., Cass, G.R.: Sources of fine organic aerosol: 5. Natural gas home appliance. Environ. Sci. Technology 27, 2736–2744 (1993a)
- Rogge, W.F., Hildemann, L.M., Mazurek, M.A., Cass, G.R., Simoneit, B.R.T.: Sources of fine organic aerosol. 2. Noncatalyst and catalyst-equipped automobiles and heavy-duty diesel trucks. Environ. Sci. Technology 27, 636–651 (1993b)
- Rogge, W.F., Hildemann, L.M., Mazurek, M.A., Cass, G.R., Simoneit, B.R.T.: Sources of fine organic aerosol. 3. Road dust, tire debris, and organometallic brake lining dust: roads as sources and sinks. Environ. Sci. Technology 27, 1892–1904 (1993c)
- Rogge, W.F., Hildemann, L.M., Mazurek, M.A., Cass, G.R., Simoneit, B.R.T.: Sources of fine organic aerosol. 6. Cigarette smoke in the urban atmosphere. Environ. Sci. Technology 28, 1375–1388 (1994)
- Rogge, W.F., Hildemann, L.M., Mazurek, M.A., Cass, G.R., Simoneit, B.R.T.: Sources of fine organic aerosol. 8. Boilers burning No. 2 distillate fuel oil. Environ. Sci. Technology 31, 2731–2737 (1997)
- Schauer, J.J., Cass, G.R.: Source apportionment of wintertime gas-phase and particle-phase air pollutants using organic compounds as tracers. Environ. Sci. Technology 34, 1821–1832 (2000)
- Schauer, J.J., Rogge, W.F., Hildemann, L.M., Mazurek, M.A., Cass, G.R., Simoneit, B.R.T.: Source apportionment of airborne particulate matter using organic compounds as tracers. Atmos. Environ. 30, 3837–3855 (1996)
- Schauer, J.J., Kleeman, M.J., Cass, G.R., Simoneit, B.T.: Measurement of emissions from air pollution sources. 1. C1 through C29 organic compounds from meat charbroiling. Environ. Sci. Technology 33, 1566–1577 (1999a)
- Schauer, J.J., Kleeman, M.J., Cass, G.R., Simoneit, B.R.T.: Measurement of emissions from air pollution sources: 2. C1 through C30 organic compounds from medium duty diesel trucks. Environ. Sci. Technology 33, 1578–1587 (1999b)
- Schauer, J.J., Kleeman, M.J., Cass, G.R., Simoneit, B.R.T.: Measurement of emissions from air pollution sources. 3. C1–C29 organic compounds from fireplace combustion of wood. Environ. Sci. Technology 35, 1716–1728 (2001)
- Schauer, C., Niessner, R., Poschl, U.: Polycyclic aromatic hydrocarbons in urban air particulate matter: decadal and seasonal trends, chemical degradation, and sampling artifacts. Environ. Sci. Technology 37, 2861–2868 (2003)
- Simoneit, B.R.T.: Characterization of organic constituents in aerosols in relation to their origin and transport: a review. Int. J. Environ. Anal. Chem. 23, 207–237 (1986)
- Wood, J.D., Enser, M.: Factors influencing fatty acids in meat and role of antioxidants in improving meat quality. Br. J. Nutr. 78(Suppl. 1), S49–S60 (1997)
- Zheng, M., Cass, G.R., Schauer, J.J., Edgerton, E.S.: Source apportionment of PM_{2.5} in the southeastern United States using solvent-extractable organic compounds as tracers. Environ. Sci. Technology 36, 2361–2371 (2002)

