Preliminary estimation of chemical compositions and emissions of particulate matters from domestic cooking in Beijing

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Abstract. Family cooking oil fume are becoming an important source of urban atmospheric fine particulate (PM2.5). In order to understand the characteristics and level of PM2.5 emission from family cooking fume. Mass concentration, chemical compositions and emission factors of PM2.5 emitted from family cooking fumes were studied based on in situ measurement, and the total amounts of PM2.5 emission from family cooking fumes in Beijing was preliminary estimated. Results showed that mass concentration of PM2.5 emitted from family cooking fumes ranged from 329.88±91.24 to 500.89±130.44 μg·m⁻³, the average mass concentration was 398.82±102.55 μg·m⁻³. Among all the measured chemical components of organic carbon (OC), water-soluble ions and element carbon (EC) account for about 51.34%-65.45%, 6.93%-14.78% and 0.73%-1.82%, respectively, other untested components account for about 19.44%-38.05%. According to the field investigation and actual measurement, the average PM2.5 emission factor was 210.57±54.15 mg·f·d⁻¹ based on household number. Combined with the 2018 statistics annual report of Beijing, the total amounts of PM2.5 emission from family cooking oil fumes in Beijing was estimated to be 553.38±142.31 t·a⁻¹ in 2018. The emission amounts has certain uncertainty, the following work should be carried out to measure the PM2.5 emission from family cooking oil fumes in different administrative regions.

1. Introduction
As people's living standards continue to improve, the pollutants discharged into the atmosphere through catering enterprises and household kitchens can no longer be underestimated. According to existing researches, the solid and liquid fumes discharged during food cooking can be condensed directly to form particulate matters, while the emitted VOCs are precursors of secondary organic aerosols[1-2]. A study covering Beijing urban areas by Wen et al.[3] found that the fine particle emissions from cooking sourceshad an equal contribution as the emissions from transportation sources to the organic particulate matters, and that the organic matters accounted for approximately 70% of the mass concentration of particulates emitted from cooking. Some studies have found that for emitted particulates of cooking sources, PM10 accounted for 97.43% of TSP mass, PM2.5 accounted for 76.91% of TSP mass[4], and PM2.5 could account for over 80% of the PM10 mass[5]. These indicate...
that the cooking fumes have become a major source of PM$_{2.5}$ in the urban atmosphere. As epidemiological studies have claimed, cooking fumes have a potential carcinogenic risk\cite{6-8}, which are in particular associated closely with the occurrence of lung cancer\cite{9-10}. Wang et al.'s\cite{11}Dongfeng–Tongji cohort study of 34,000 subjects found that the longer the daily cooking time, especially if exceeding 2 h, the higher the risk of developing lung cancer. Besides, the effects of oil fumes on human genotoxicity and immunity are also non-negligible\cite{12-13}.

On the basis of the above research background, abundant achievements have been made at home and abroad concerning the emission characteristics of organized cooking emission sources, such as street and mall restaurants\cite{14-19}. In contrast, there are few reports on the cooking fume emissions from residents' domestic life. Household cooking, as an unorganized emission source, is scattered among huge numbers of households in cities. Given its differing emission characteristics and management methods from those of catering companies, people have limited understanding of its actual existence, activity level and emission quantity. Liu et al.\cite{20} estimated the cooking fume emissions in Beijing based on the literature survey data, and concluded that the PM$_{2.5}$ emissions from household cooking accounted for 11.74% of the total PM$_{2.5}$ emissions from all cooking sources. Wu et al.\cite{21} derived PM$_{2.5}$ emission factors benchmarked against the dining frequency, oil consumption and cooking range based on the literature data. They found that the domestic cooking contributed 74%, 81% and 75% of all cooking activities in Changchun. Through literature research and actual testing, Qin et al.\cite{22} proposed emission factors benchmarked against population and cooking oil, who concluded that the household cooking emissions in Chengdu accounted for 50.68% and 60.77% of the total PM$_{2.5}$ emissions from cooking sources, respectively. From the above studies, it is clear that the fume emissions from domestic cooking have a great contribution to the PM$_{2.5}$ emissions of cooking sources. Additionally, accounting method and choice of emission factor under different accounting benchmarks are crucial to the computation of fume PM$_{2.5}$ emissions. Thus, to obtain accurate local emission factors through a combination of actual measurement and field survey, it is critical to estimate the PM$_{2.5}$ emissions from domestic cooking scientifically.

Three communities in key areas of Beijing's Chaoyang District were selected for study herein. The domestic cooking fume samples were collected via modified exhaust vents, and the concentration, chemical components and emission factors of PM$_{2.5}$ in the fumes were determined. On these bases, the activity level information in the Beijing Statistical Yearbook was utilized to estimate the total annual emissions of PM$_{2.5}$ from domestic cooking. In this study, the emissions characteristics and levels of PM$_{2.5}$ from domestic cooking were derived through actual measurement, which have important referential value for Beijing in adopting comprehensive, reasonable control technology and policy measures to manage the domestic cooking fumes.

2. Materials and methods

2.1. Design of sampling system

Domestic cooking fumes are discharged in a way different from the commercial cooking fumes. There is no fan or purification system at the fume exhaust vents on the top of residential buildings, and the relevant exhaust flow rate is constituted mainly by the hot air temperature difference in kitchen and the power of range hood. Both the flow rate and concentration of exhaust are extremely unstable, with huge intra-day and inter-day differences, which are thus detrimental to the sample collection and subsequent estimation of emissions. To assess the level of particulate matter emissions from domestic cooking accurately, we modified the fume exhaust vents in this study by installing an induced draft fan at the rear. By doing so, a continuous and stable discharge flow can be attained, thereby lowering the experimental errors. In figure 1, the design schematic of the sampling system is displayed, as well as a photo of sampling site.
2.2. Design of sampling scheme

The object of this study is domestic cooking fumes. Compared to a commercial restaurant, the duration, frequency and intensity of cooking by a single household are all considerably smaller, and the level of pollutant emission concentration is also far lower. Besides, since the fume exhaust vent is shared by multiple households, the emission period is rather scattered due to the varying life styles of households. In addition, Beijing is a haze-prone city with high background values of atmospheric particulate matters. Therefore, removal of background interference is particularly important during the detection of particulate matters from cooking fumes herein. Based on the above considerations, a 24 h all-weather sampling scheme was designed for collection of particulate matters. It not only enables acquisition of more cumulative mass of particulate matters in a single sample, but also eliminates the time discretization error caused by discontinuous emissions. Besides, to minimize the interference resulting from the ambient air background, during collection of the fume particulate matter samples, the particulate matters from the indoor bathroom vents of the same residential building were also collected with the same sampling system, which serve as the background blank. In figure 2, the principle of sampling scheme is illustrated.
2.3. Sample collection
Prior to the sample collection, the quartz filter membrane (φ90 mm, Whatman, USA) was placed in a muffle furnace and baked at 550 °C for 4 h, so as to remove the residual organics and other organic impurities on the membrane. All metal products, such as the sampler brackets and the tweezers for sampling, were ultrasonically cleaned with dichloromethane and acetone prior to use. Both before and after weighing, the filter membrane was equilibrated in a constant temperature and humidity chamber for more than 24 h, and the weighing conditions before and after sampling were kept consistent. Afterwards, the weighed filter membrane was placed in a special capsule, which was then wrapped with a sealing film and additional aluminum foil, packed into a sealed bag and stored in a -18 °C refrigerator for later use.

As a core district of Beijing, Chaoyang District has the largest size of population and number of commercial dwellings in Beijing. Accordingly, the selection of residential buildings in Chaoyang District as the research object has a high representativeness. Within this district, one residential building was chosen each from three communities, namely the ZaoyingBeili, Chaoyang Park Xili and AnhuiBeili. Within each residential building, two cooking fume exhaust vents (A and B) were selected. Meanwhile, the residential indoor air was sampled separately at the corresponding bathroom vents to serve as the background blank samples. A medium flow particulate sampler (100 L·min⁻¹, Laoying, Qingdao) was used to collect the PM₂.₅ samples. The sampling time was 24 h, with an error range of ≤0.5 h. Each community was monitored continuously for 14 d. To facilitate subsequent data statistics and analysis, during the field monitoring, the architectural structure of monitored buildings and the daily habits of residents were also investigated in situ.

2.4. Sample collection

2.4.1. Determination of PM₂.₅ mass concentration. The quartz filter membrane was weighed three
times before and after sampling on a 100,000th electronic balance (CP 225D, Sartorius, Germany), and then averaged. Afterwards, the filter membrane was placed for additional 24 h in the same conditions, and then weighed and averaged. The mean difference between the two operations was required to be <0.01 mg. Otherwise, the procedure was repeated until the requirements were fulfilled. The mass concentration of PM$_{2.5}$ in cooking fumes was computed according to the collected mass and the actually sampled volume.

2.4.2. Determination of OC and EC in PM$_{2.5}$. The thermal-optical reflectance (TOR) carbon analyzer (Model 2001A, DRI, USA) was used to determine the organic carbon (OC) and elemental carbon (EC) in PM$_{2.5}$. From the quartz filter membrane, 0.53 cm$^2$ circular sample was cut with a sampler, which was then heated in an anaerobic pure helium environment by the IMPROVE A protocol, thereby releasing different components of OCs (OC1, OC2, OC3 and OC4) at 140 °C, 280 °C, 480 °C and 580 °C, respectively. Next, the sample heating was continued in a helium environment with 2% oxygen, thereby releasing different components of ECs (EC1, EC2, EC3) at 580 °C, 740 °C and 840 °C, respectively. The carbon-containing compounds evaporated at each of the above temperature gradients were converted to CO$_2$ by catalytic oxidation of MnO$_2$, and then converted to CH$_4$ by catalytic reduction of Ni, followed by quantitative detection with a flame ionization detector (FID). During the anaerobic heating of sample, some OCs could be carbonized to form pyrolytic carbon (OPC), which were contained in EC1, thereby leading to low measurement results of OC and high measurement results of EC. To correct the measurement errors resulting from carbide formation, a 633 nm He-Ne laser was used to monitor the changes in reflected light intensity of filter paper throughout the measurement process. The time points at which the reflected signal left and returned to the baseline position were recorded. The carbons detected within this period of time could be deducted from EC, and then re-corrected into OC. The final OC was defined as OC1+OC2+OC3+OC4+OPC, while the final EC was defined as EC1+EC2+EC3-OPC. The limits for instrumental detection of OC and EC were set at 0.82 μg·cm$^{-2}$ and 0.20 μg·cm$^{-2}$, respectively.

2.4.3. Determination of water-soluble anions and cations in PM$_{2.5}$. One quarter of the quartz membrane was immersed in 15 mL of deionized water (18.2 MΩ), and ultrasonically extracted for 60 min. After filtering the extract through a 0.45 μm microporous membrane, the ICS2000 ion chromatograph (Dionex, USA) was utilized to analyze four kinds of anions (F$^-$, Cl$^-$, NO$_3^-$ and SO$_4^{2-}$) and five kinds of cations (Na$^+$, NH$_4^+$, K$^+$, Mg$^{2+}$ and Ca$^{2+}$) in the sample. For anion analysis, AS11-HC column (250 mm x 4 mm) was used, and the eluent was 15 mmol·L$^{-1}$ KOH. Meanwhile, CS12-A column (250 mm x 4 mm) was used for cation analysis, and the eluent was 20 mmol·L$^{-1}$ methanesulfonic acid. For various ions (F$^-$, Cl$^-$, NO$_3^-$, SO$_4^{2-}$, Na$^+$, NH$_4^+$, K$^+$, Mg$^{2+}$ and Ca$^{2+}$), the detection limits were set at 0.0011, 0.0025, 0.0028, 0.0036, 0.0015, 0.0018, 0.0023, 0.0021 and 0.0045 mg·L$^{-1}$, respectively.

2.5. Quality control and assurance

All experimental steps in this study were performed under strict quality control. Prior to each sample collection, the sampling system was washed, wiped, checked for air tightness and calibrated for flow rate. The system was run blank before the measurement of OCs and ECs in sample. If the blank was beyond the acceptable limit (TC 0.50 μg C), the system would be baked again until the system blank was beneath the limit value. Additionally, a system stability test (trimodal detection) was performed once each at the beginning and end of the OC and EC analyses every day for quality assessment. The deviation should not exceed 5%, or the sample analysis could not be initiated. During detection of water-soluble ions, quality control was executed in strict accordance with the HJ800-2016 and HJ799-2016.

3. Results and discussion

3.1. Flow rate evaluation of modified exhaust system
To ensure that the samples were collected in the same flow rate condition, an induced draft fan was installed at the rear of sampling channel. In figure 3, the exhaust flow rates of sampling port in any one hour are depicted. Clearly, at the modified exhaust vents that were installed with an induced draft fan, the emission velocity of fumes is stabilized at around 7 m·s⁻¹ in any one hour. Since the velocity did not change with the peak and trough cooking periods, it could meet the sampling requirements for particulates from organized emissions.

![Figure 3. Exhaust flow rate of sampling port in any one hour.](image)

3.2. Emission characteristics of PM₂.⁵ from domestic cooking

From March 8, 2019 to April 28, 2019, a total of 42 sets of valid fume particulate samples were collected from domestic cooking, which totaled 168 samples. The blank background values were already subtracted from all the experimental data. As shown in figure 4, the mass concentrations of PM₂.⁵ from family cooking collected at different sites of residential communities during the sampling period ranged between (329.88±91.24)-(500.89±130.44) μg·m⁻³, with a mean of 398.82±102.55 μg·m⁻³. The mass concentrations of OCs ranged between (192.63±14.25)-(257.85±48.99) μg·m⁻³, with a mean of 227.51±41.42 μg·m⁻³, which accounted for approximately 51.34%-65.45% of the sample mass concentration. ECs exhibited relatively small contributions, accounting for 0.73%-1.82% only. Water-soluble ions accounted for about 6.93%-14.78%, while other unmeasured components accounted for about 19.44%-38.05% (see figure 5). Clearly, carbon-containing components, especially OCs, were a major part of PM₂.⁵ in domestic cooking fumes.

![Figure 4. Mass concentrations of PM₂.⁵ and their chemical compositions from family cooking in different sites.](image)
According to figure 6, among the water-soluble ionic components, the mass fractions of $SO_4^{2-}$, $Cl^-$, $Na^+$ and $NO_3^-$ were generally high, which were 17.60-39.11%, 16.14-31.19%, 9.59-15.87% and 7.14-15.49%, respectively. The total proportion of the four ions could reach 65.96-84.54% of entire water-soluble ions. As studies have demonstrated, $Na^+$ and $Cl^-$ come primarily from salt, and there are other components like $Na_2SO_4$ and $NaNO_3$[3]; $SO_4^{2-}$ and $F^-$ are mainly from drinking water[15]; while other ions are mainly from food ingredients and cooking oil[23]. In addition, different cooking methods, fuels and the use of element-rich food ingredients also constitute important factors affecting the PM$_{2.5}$ ion components of fume emissions[24].

3.3. Determination of emission factors

Emission factor method, as the most popular way of accounting emissions, is also the principal method developed by China's emission inventory of cooking fumes. The emission factors vary greatly among accounting methods. After comprehensively considering the accuracy of activity level data and the
difficulty of data acquisition, we finalized the number of households as the benchmark. And the emission factor was defined as the PM$_{2.5}$ emissions from cooking fumes per household per day. According to the aforementioned sampling scheme, the average diurnal emission of PM$_{2.5}$ per household was calculated as:

\[
\frac{M_{\text{fume}} - M_{\text{blank}}}{V_{\text{collection}} \times N} \times V_{\text{emissions}}
\]

Where $M_{\text{fume}}$ denotes the mass of PM$_{2.5}$ collected at the flue vent, g; $M_{\text{blank}}$ denotes the mass of PM$_{2.5}$ collected at the control vent, g; $V_{\text{emissions}}$ represents the flue flow rate, m$^3$·h$^{-1}$; $V_{\text{collection}}$ represents the sampling flow rate, L·min$^{-1}$; and $N$ is the number of households corresponding to each exhaust vent.

In figure 7, the PM$_{2.5}$ emissions generated by cooking fumes per day in six flues are displayed, at a flue flow rate of 792 m$^3$·h$^{-1}$ and a sampling flow rate of 100 L·min$^{-1}$. As can be seen, among the three selected communities, the highest emission value of 9.52±2.48 g·d$^{-1}$ was found at the vent A of building 2 in Chaoyang Park Xili, whereas the lowest emission value of 6.27±1.73 g·d$^{-1}$ was noted at the vent B of building 2 in Anhui Beili. There were certain inter-building differences in the emissions. All the residential buildings selected herein were 18-floor tower structures. According to the field survey, each exhaust vent corresponded to 36 households. Hence, the average diurnal PM$_{2.5}$ emissions from cooking fumes per household was calculated by formula (1), whose value was 210.57±54.15 mg·household·d$^{-1}$.

![Figure 7. Average diurnal emission of PM$_{2.5}$ from family cooking fume in different sites.](image_url)

3.4. Estimation of emissions

In the Beijing Statistical Yearbook 2018, the total population of Beijing was 21 million, the total number of households was approximately 7.2 million, and the population of Chaoyang District was 17.10% of the entire Beijing population. Assuming each household had three people, then the number of households in Chaoyang District was about 1.2 million. Based on the emission factor measured above, the annual emissions of PM$_{2.5}$ from domestic cooking in Chaoyang District was derived to be approximately 92.23±23.72 t·a$^{-1}$. The total emissions of PM$_{2.5}$ from domestic cooking in Beijing was estimated while ignoring the differences in average diurnal PM$_{2.5}$ emissions from domestic cooking per households between districts, which was found to be 553.38±142.31 t·a$^{-1}$. 
Table 1. PM$_{2.5}$ emission amounts of cooking sources in Chinese cities.

| Study site | Study year | Emission type       | Total emissions/ t·a$^{-1}$ | Reference |
|------------|------------|---------------------|----------------------------|-----------|
| Beijing    | 2005       | Commercial catering | 2100                       | [3]       |
| Shanghai   | 2014       | Commercial catering | 681-946                    | [25]      |
| Guangzhou  | 2011       | Commercial catering | 1248                       | [26]      |
| Nanjing    | 2014       | Commercial catering | 3160                       | [27]      |
| Changchun  | 2018       | Household dining    | 259-571                    | [21]      |
| Chengdu    | 2013       | Household dining    | 3190                       | [28]      |
| Chengdu    | 2014       | Household dining    | 1347-1817                  | [22]      |
| Beijing    | 2010       | Domestic cooking fume | 1528                      | [20]      |
| Beijing    | 2019       | Domestic cooking fume | 553.38±142.31             | Present study |

Comparison with the PM$_{2.5}$ emissions from cooking fumes in Chinese cities (see table 1) found larger differences in the emission values between cities, as well as among studies for the same cities. The probable causes were as follows: The dining habits varied region by region; and the method used for measuring emissions differed from the accounting benchmark. Moreover, some studies utilized non-local emission factor data, thus leading to certain biases in emissions estimates. The total annual PM$_{2.5}$ emissions from domestic cooking in Beijing estimated in this study was less than that reported by Liu et al.[20]. Regarding the possible reasons, firstly, the estimation method used herein was based on the actual measurements of household cooking fumes, which were inevitably less than the literature research-based theoretical statistics. Secondly, this study targeted the emission of particulate matters generated from cooking fumes, thus eliminating the effect of background blank values in the residential buildings. Due to a combination of these two factors, the results of the present experiment were slightly lower, but were closer to the real emission values of domestic cooking fumes.

3.5. Uncertainty analysis
Due to the inadequacy of data representativeness and the limitations of estimation method, uncertainties were unavoidable during the estimation of PM$_{2.5}$ emissions from domestic cooking. In this study, the uncertainties came from three aspects: (1) Determination of emission factors. This study targeted the PM$_{2.5}$ generated from cooking fumes, and selected the bathroom vent as the background blank value for subtraction. Given the varying lifestyles, dining habits and indoor environments among residents, the measured data had certain limitations. (2) Determination of number of households. The population size of Beijing in this study came from the Beijing Statistical Yearbook. The number of households allocated according to the population size was somewhat uncertain. (3) Selection of sampling sites. This study mainly chose tower buildings from the residential communities in Chaoyang District's key areas. Given the lack of different districts and architectural structure types, the data were insufficiently representative.

4. Conclusions
During the sampling period, the mass concentrations of PM$_{2.5}$ from cooking fumes collected at various residential communities range between (329.88±91.24)-(500.89±130.44) μg m$^{-3}$, with a mean of 398.82±102.55 μg m$^{-3}$. The chemical components, sorted in a descending order of mass concentration, are OCs, water-soluble ions and ECs, which account for 51.34-65.45%, 6.93-14.78% and 0.73-1.82% of PM$_{2.5}$ mass concentration, respectively. There are also unmeasured components, which account for approximately 19.44-38.05%. Among the water-soluble ions, SO$_4^{2-}$, Cl$^-$, Na$^+$ and NO$_3^-$ occupy relatively higher proportions, with a total proportion reaching up to 65.96-84.54% of entire water-soluble ions.
After actual measurement combined with field survey, the PM$_{2.5}$ emission factor for domestic cooking, which is benchmarked against the number of households, is derived to be $210.57 \pm 54.15$ mg household$^{-1}$d$^{-1}$. On this basis, the activity level information in the Beijing Statistical Yearbook 2018 is utilized to estimate the total emissions of PM$_{2.5}$ from domestic cooking fumes in Beijing, whose value is approximately $553.38 \pm 142.31$ t a$^{-1}$. Some uncertainties are inevitable in the level of domestic cooking fume emissions measured in this study. In the subsequent works, actual measurement of fume emissions should be carried out on typical residential communities with different building structures in various regions. Moreover, quantitative analysis of emission source uncertainties should be strengthened, in order to further improve the analyses on the quantity and characteristics of PM$_{2.5}$ emissions from domestic cooking.

Lastly, with the transition of energy structure and the implementation of policies like the outward manufacturer relocation in Beijing, the contribution of cooking fumes to atmospheric PM$_{2.5}$ will become prominent. At present, the cooking source emissions generally do not include the emissions of domestic cooking fumes. From the perspectives of control of pollution sources and their proportions, it is advisable to list the emissions of domestic cooking fumes in the PM$_{2.5}$ emission inventory of cooking sources for control.

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