Emissions of Volatile Organic Compounds (VOCs) from Cooking and their Speciation: A Case Study for Shanghai with Implications for China

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Abstract: Cooking emissions are an important source of ambient volatile organic compounds (VOCs), which are deleterious to air quality, climate and human health. These emissions are especially of great interest in large cities of East and Southeast Asia, concerning its significant loading and impacts on climate and human health. We conducted a case study in which VOC emissions from kitchen extraction stacks have been sampled in total 57 times in the Megacity Shanghai. To obtain a representative dataset of cooking VOC emissions, focuses have been given to cuisine types, including restaurants of seven common, canteens, and family kitchens. VOC species profiles and their chemical reactivities have been determined. The results showed that alkane and oxygenated VOCs (O-VOCs) dominate the VOC cooking emissions, with contributions of 13.3-65.9% and , respectively. However, the VOCs with the largest ozone formation potential (OFP) and secondary organic aerosol potential (SOAP) were from the alkene and aromatic categories, accounting for 6.8-97.0% and 73.8-98.0%, respectively. Barbequing has the most potential of hazardous health effect due to its relatively higher emissions of acetaldehyde, hexanal, and acrolein. Methodologies for calculating VOC emission factors (*EF*) for restaurants counting as VOCs emitted per person (EF_{person}), per kitchen stove ($EF_{kitchen\ stove}$) and per hour (EF_{hour}) are developed and discussed. Methodologies for deriving VOC emission inventories (S) from restaurants are further defined and discussed based on two categories: cuisine types (S_{type}) and restaurant scales (S_{scale}). The range of S_{type} and S_{scale} are 4124.33-7818.04 t/year and 1355.11-2402.21t/year, respectively. We also reported that the S_{type} and S_{scale} for 100,000 people are 17.07-32.36t/year and 5.61-9.95t/year in Shanghai, respectively. Based on Environmental Kuznets Curve, the annual total amounts of VOCs emissions from catering industry in different provinces in China have been estimated as well. For the total amount of VOCs emissions, Shangdong and Guangdong provinces and whole China reach up to 5680.53 t/year, 6122.43 t/year, and 66244.59 t/year, respectively. In addition, we suggest that large and medium-scale restaurants should be regarded as the most important factors with respect to regulation of VOCs.

Keyword: Cooking emissions; Volatile organic compounds; Emission Inventory; Emission factors; Restaurant scales

INTRODUCTION

Volatile organic compounds (VOCs), as important precursors of ozone and secondary organic aerosols (SOAs), are critical for the formation of photochemical smog and fine particulate matter in the atmosphere(Atkinson, 2000;Volkamer et al., 2006;Kroll et al., 2006). These deleterious compounds have a significant impact with respect to climate change and air quality, and cause adverse health effects on human beings (Fiore et al., 2008;Massolo et al., 2010). The role of VOCs in terms of air quality in China and Southeast Asia has becoming more and more serious, owing to the unsound emission standards and waste disposal measures. Urban areas among a number of cities in these regions are suffering from haze, and SOAs have been proven to be one major factor (Huang et al., 2014;Guo and Lakshmikantham, 2014). In addition, the problem of ozone pollution is becoming more and more serious in East and Southeast Asia (Wang et al., 2017a). There have been already a number of studies on cataloging VOC emission inventories originating from vehicles, biomass burning and industrial processes, especially in China (Bo et al., 2008;Guo et al., 2007;Huang et al., 2011a;Liu et al., 2005;Yin et al., 2015;Zheng et al., 2017). As one of the significant source impacting urban air quality and human health, only a number of studies compare emissions from different cooking processes, but not characterize how cooking emissions enter into the ambient urban atmosphere (Wang et al., 2017b). In China and other countries of Southeast Asia, people usually employ often high temperature oil for frying food on a daily basis. Over 300 kinds of reaction products have the potential to be released during this cooking process (Wang et al., 2017a). One hotspot for air pollution is for example Eastern China because of its high population density and rapid urbanization.

For this case study, Shanghai was chosen as the largest city in this area. Here, the restaurant business is well developed in terms of both scale and variety. In 2012, the total number (2012) of registered restaurants in Shanghai have been 36,692. Characterizing VOC emissions and their reactivity profiles from such a large commercial sector is thus an urgent issue, which has to be investigated and understood. Exploring the species profiles of VOCs produced from cooking in Shanghai’s urban area and creating emission inventories will allow for meaningful regulatory policy. Furthermore, as a result of the complexities of quantifying VOC emissions from various cuisine types and the unexpected randomness of customer demands, the methodologies for building up inventories for VOC emissions arising from urban cooking and their related emission factors have not been well established yet.

Motivated by this urgent need, this study represents the initial foray into establishing a VOC emissions inventory that represents multiple residential and commercial kitchens in Shanghai. A total of 57 rounds of in-situ measurements of VOC emissions from the extraction stacks of restaurants for seven cuisine types in Shanghai, including canteens and family kitchens, were investigated. The aim was to identify the similarities and differences between VOC compositions and their chemical reactivity among the different types of urban kitchens, and propose methodologies for deriving VOC emission factors and inventories. All restaurants were compared by employing a classification scheme based on cuisine types and restaurant scales. For each classification, emissions per person, per kitchen stove, and per hour, as well as which emission factors are most recommended, are discussed. The conclusions provides the foundation for building a continuing body of statistical knowledge and methodologies that can be used in calculating emission factors, inventories, and total annual amount for other cities and nations, as well as for assessing the impact of cooking emissions on urban atmosphere and human health.

MATERIALS AND METHODS

Sampling Methodology. Restaurants of seven cuisine types were selected for sampling at their emission extraction stacks, including: Authentic Shanghai cuisine, Shaoxing cuisine, Cantonese cuisine, Western fast food, Sichuan and Hunan cuisine, Fried food and Barbecue. Canteens and Family kitchens were also investigated. The sampling time was chosen to be during lunch (11:30~13:30) or dinner (16:30~18:30) periods. Two to three samples were collected continuously for each round of measurement. Detailed information is given in Table SI1.

The sampling point was set at 0.5 m above the extraction stack. For small scale restaurants and street food vendors without smoke channels, the sampling point was about 0.5 m above the operation area containing the cooking appliances. 3.2L SUMMA canisters, pipes and connections were cleaned several times with ENTECH equipment before each measurement, and followed with vacuum backup. Each canister was connected with a Teflon filter to remove particulate matter and moisture during sampling. Real-time monitoring of non-methane hydrocarbons (NMHCs) was conducted using a J.U.M 3-900 heated FID total hydrocarbon analyzer. The setup is shown in Figure SI1.

VOCs Analysis. The collected samples were analyzed using gas chromatography-mass spectrometry (GC-MS, Agilent, GC model 7820A, MSD model 5977E). Photochemical Assessment Monitoring Stations (PAMs) were adopted to quantitatively determine 99 types of VOC species. All samples went through the automatic sampler for precooling enrichment treatments prior to entering the GC-MS. The precooling concentrator extracted a certain amount of samples by trapping them into a $^1/_4$ inch liquid nitrogen trap. After the water and CO_2 was removed, the samples were separated by GC, and then entered the MS to be spectrometrically analyzed. The temperature program initiated with a 3 min isothermal period at –35℃, followed by a ramp to 220℃ at a rate of 6℃/min, and remained at 220℃ for 6 min. The carrier gas was helium. Target compounds were identified using their chromatographic retention times and mass spectra, and the concentrations of target compounds were calculated using internal standard method. The detection limit was from a fraction of μg/m^3 to over ten μg/m^3(Jia et al., 2009;Qiao et al., 2012). VOC species were identified by their retention time and mass spectra. A commercial standard gas (Spectra, USA) containing PAMS (Photochemical Assessment Monitoring System), O-VOC, and x-VOC was used to identify compounds and confirm their retention times. 99 species including 29 alkanes, 11 alkenes, 16 aromatics, 14 O-VOC, 28 x-VOC and acetylene were identified in this study.

RESULTS AND DISCUSSIONS

Speciation of VOCs Arising from Cooking Emissions

Cooking emissions are generated via intensive chemical reactions occurring with edible oil or food under high temperatures by three major pathways: 1) thermal oxidation and decomposition of the lipid; 2) Maillard reaction of some chemical species; 3) secondary reaction of the intermediates or final products (Kleekayai et al., 2016). VOCs mainly come from heated oils and fatty acids. The former is related to triglycerides, of which the double bond location and the fracture location cause generation of different hydroxyl species and further leads to decomposition into alkanes and alkenes (Choe and Min, 2006). The profiles of 99 VOC species were obtained, as listed in Table SI2. Normalization was carried out in order to calculate their mass concentrations.

Figure 1 reveals that alkanes were the major VOC pollutant, a fact which can be attributed to the large consumption of peanut oil in Shanghai (He et al., 2013). Incomplete combustion of fats derived from meats is a secondary explanation (Hildemann et al., 1991;Rogge et al., 1991). Fugitive emissions from liquefied petroleum gas (LPG) and natural gas (NG), which are usually used as the fuel source for cooking, was another added source of alkanes, leading to the increased prevalence of propane, n-butane, and i-butane. Aldehydes, generated by shallow frying of food, also dominated as a result of the decomposition of fatty acids instead of heated oil (Wood et al., 2004), and were also major species in most cuisine types.

Figure 1.

Generally, the investigated cuisine types can be classified into six categories. *1) Canteen, Authentic Shanghai cuisine and Cantonese cuisine.* The proportion of alkanes was the largest, followed by alkenes and O-VOCs. The main components of the alkanes were ethane and propane for canteen and Authentic Shanghai cuisines. C2, C8 and C3 alkanes were the greatest contributors with respect to Cantonese cuisines. *2) Shaoxing cuisine.* C2 to C5 alkanes were the largest contributors. Acetylene was predominant as well. A greater quantity of alkenes and O-VOCs were observed, which was possibly due to the use of rice wine and fresh ingredients adopted for stews. The abnormally high acetylene concentration might be a consequence of the equipment of the facilities. *3) Western fast food, Sichuan and Hunan cuisine.* C3~C6 and C2~C6 alkanes were the major O-VOC contributors for each restaurant type, respectively. Acrolein, n-hexaldehyde and acetone were the dominant contributors. Acrolein is only generated from edible oils, hence the enhanced consumption of oil is likely to be the reason for the relatively greater O-VOC production. An abundance of acetone usually exists in vegetables and volatilizes during boiling. One such example are onions(Huang et al., 2011b), which are used very often for these two cuisine types, and are likely a major source for acetone. Evaporative loss of impurities in fuels is a reason for the significant increase of aromatic and X-VOCs (Huang et al., 2011b). *4) Fried food.* Alkanes and O-VOCs contributed to over 97% of the total VOCs, owing to meat-derived fats and large quantities of oil, respectively. The dominant species of alkanes were 2, 2, 4-trimethylpentane and n-pentane. The main components of O-VOCs were hexanal, pentanal and acetaldehyde. *5) Barbecue.* Alkanes contributed here over 83%, as a result of the consumption of large amounts of fat and the adoption of charcoal as a fuel. The main alkane compounds were 2, 2, 4-trimethylpentane and 2 - methylhexane. *6) Family kitchen.* Alkanes and O-VOCs were 44.7±1.5% and 32±0.6%, respectively. 2, 2, 4-trimethylpentane and 2 - methylhexane accounted for the largest percentage for the alkanes. Hexanal, acetaldehyde and acetone were the main substances of the O-VOCs.

Figure 2 compares VOC compositions obtained from this study with other studies. Generally, similar results were obtained among all of the different studies, and alkanes were the dominant contributor for all reports. The observed discrepancies can be attributed to differences in restaurant scales, ambient pollutant concentrations and emission sources.

Figure 2.

Ozone Formation Potential of VOCs. OFP was calculated by taking into account VOC source profiles together with the maximum incremental reactivity (MIR) of each species (Carter, 1994). Normalized percentages of OFP for each category of VOCs for all cuisine types are shown in Figure 3. The average MIR for VOCs from different cuisine types was calculated as the ratio of total OFP to VOC concentration, which can be thought of as the average OFP per unit mass of VOC emission, as given in Figure 3.

Figure 3.

Figure 3 reveals that the top three contributors to OFP were alkenes, O-VOCs and alkanes for Canteen, Authentic Shanghai cuisine, Shaoxing cuisine and Cantonese cuisine, respectively. The chemical reactivity of ethylene and acetaldehyde accounted for 46.9±3.2–69.2±12.5% and 8.0±1.4–11.7±3.5%, respectively. The largest contributors were O-VOCs and aromatics for Western fast food, Sichuan and Hunan cuisine and fried food. Acetaldehyde and hexanal accounted for 20.5±1.1–35.2±2.9% and 11.4±2.3–24.1±9.4% of the total OFP, respectively. With respect to barbeque, alkenes contributed to 56.0±12.5% of total OFP. The major contributing species were acrylic acid (25.6±4.6%), isooctane (25.6±4.9%) and ethylene (19.0±7.3%). Alkenes (C2–C4) were also the main source of chemical reactivity for Fried food, and isooctane was the largest contributor in this category as well. O-VOCs and alkenes contributed 53.3±12.6% and 29.9±3.4% to the total OFP for family kitchens, respectively. Acetaldehyde (24.2±3.5%), n-hexanal (10.9±4.8%), propylene (10.0±2.7%) and ethane (9.3±3.5%) were the largest contributors. It was also concluded by the data shown in Figure 3 that the average MIR of VOCs from cooking emissions ranged from $3.0\times10^{-12}\cdot\text{cm}^3\cdot\text{molecule}^{-1}\cdot\text{s}^{-1}$ to $11.5\times10^{-12}\cdot\text{cm}^3\cdot\text{molecule}^{-1}\cdot\text{s}^{-1}$, among which, Western fast food, Sichuan and Hunan cuisine, and family kitchens showed the highest MIR.

SOA Formation Potential of VOCs. SOA formation potential (SOAP) represents the propensity for an organic compound to form secondary organic aerosols, when that compound is emitted to the ambient atmosphere. The value is generally reported relative to the secondary organic aerosol formations of toluene, when an identical mass concentration of the species of interest is emitted into the atmosphere(Derwent et al., 2010;Johnson et al., 2006;Kleindienst et al., 2007;Hu et al., 2008), as described by equation (1):

$$SOAP_i = \frac{Increment\ in\ SOA\ mass\ concentration\ with\ species;\ i}{Increment\ in\ SOA\ with\ toluene} \times 100 \qquad (1)$$

SOAP mass-weighted contributions(Derwent et al., 2010) of each VOC category is shown in FigureSI2. Aromatics accounted for 75.34±15.35–98.14±19.54% of the total. The largest contributor was toluene. Although VOCs with low carbon numbers dominated, their contribution to SOA formation can be neglected. The saturated vapor pressures for oxidizing VOCs with low carbon numbers are too high, such that these VOCs do not tend to condense into aerosol phases(Derwent et al., 2010).

VOC Emission Factors. Emission factors of VOCs and NMHCs related to per person (EF_{person}, g/person), per kitchen stove ($EF_{kitchen\ stove}$, g/h·stove), and per hour (EF_{hour}, g/h) were investigated. Background VOC concentrations for each individual measurement were subtracted prior to performing the calculations. Emission factors for VOCs and NMHCs were calculated according to equation (2–4), respectively:

$$EF_{person} = \frac{\sum_i VOC_i \times F \times 10^6}{P} \quad \text{or} \quad EF_{person} = \frac{NMHC \times F \times 10^6}{P} \tag{2}$$

$$EF_{kitchen\ stove} = \frac{\sum_i VOC_i \times F \times 10^6}{N} \quad \text{or} \quad EF_{kitchen\ stove} = \frac{NMHC \times F \times 10^6}{N} \tag{3}$$

$$EF_{hour} = \sum_i VOC_i \times F \times 10^6 \quad \text{or} \quad EF_{hour} = NMHC \times F \times 10^6 \tag{4}$$

where VOC_i is the mass concentration of species i, μg/m^3. $NMHC$ is the mass concentration of NMHC, μg/m^3. F is the flow rate, m^3/h. P is the hourly number of customers, person/h. N is the number of kitchen stoves in each restaurant. Based on the information of the number of people and kitchen stoves collected during sampling (Table SI3), the calculated three types of emission factors for each cuisine type are given in Table 1.

Table 1.

According to the Shanghai Municipal Food and Drug Administration, restaurants can be classified into extra-large, large, medium or small scales based on the amount of area occupied and the number of seats(FDA, 2011). Emission factors derived by considering restaurant scales are given in Table 2. Emission factors for both large and medium-sized restaurants were the most significant, and so these restaurant sizes should be the focus for management control.

Table 2.

The variances in Table 2 were generally less than in Table 1, especially for authentic Shanghai and Cantonese cuisines, which taken together accounted for the major portion of large and medium scale restaurants. This result indicates that pollutant emissions entering the ambient atmosphere are mainly determined by restaurant scales. Hence, emission factors based on restaurant scales are recommended for estimating VOCs produced from urban cooking activity. Furthermore, with respect to the emission factors of per person, per kitchen stove and per hour, whether all kitchen stoves were turned on and whether the kitchens sampled in the study are enough to provide an accurate representation of the entire population are questions, which still need to be addressed. Therefore, EF_{hour} is recommended as long as the statistical data of the restaurants and the emission concentrations monitored from the extraction stacks of each restaurant is accurate.

VOC Emission Inventories Based on Cuisine Types. Two categories of emission inventories were included that took into account cuisine types and restaurant scales. According to the previously defined three types of emission factors, the first methodology based on EF_{person} was calculated as equation (5):

$$S_{person-type} = 52 \times \sum_j\left(\sum_i(Q \times y_i \times e) \times x_j \times EF_{person\ i}\right) + 52 \times \sum_t^2\left((Q \times 21 - \left(\sum_i(Q \times y_i \times e)\right) \times z_t \times EF_{person\ t}\right) \quad (5)$$

where Q is the population of Shanghai, which was 24,152,700 by the end of 2015; y_i is the percentage of the Shanghai population dining in each restaurant type, %; e is the number of meals per week in restaurants for Shanghai residents; z_t is the percentage of dining frequency taking place in a canteen or at home; x_j is the percentage of customer preferences by cuisine type, %.

According to a survey conducted by the Chinese Cuisine Association for people dining in restaurants, among all the respondents, 6.2% dined four times a week, 51.1% dined 2–3 times a week, 38.8% dined once or less per week, and 3.9% dined every single day(CCA, 2015), as shown in Figure 4(A). Then we obtained the Shanghai population dining distributions based on customer dietary preferences(CCA, 2015), as given by Figure 4(B) and (C). We assumed a third of the remaining population dine in canteens, and two-thirds eat at home. According to equation (5), an annual VOC emissions from cooking in Shanghai of 7818.04±254.32 t Yr^{-1} was obtained, as shown in Figure 4(D). The annual NMHC was found to be 15226.85±3755.12 t Yr^{-1}.

Figure 4.

The second methodology which is based on $EF_{kitchen\ stove}$ is described by equation (6):

$$S_{kitchen\ stove-type} = 365 \times \sum_i(EF_{kitchen\ stove} \times t \times Na \times a) + EF_{kitchen\ stove} \times Nc \times t \times 365 \quad (6)$$

where Na is the number of each cuisine type in Shanghai; a is the number of kitchen stoves for each cuisine type; Nc is the number of families in Shanghai. Household emission statistics and the sixth national census showed that the number of households in Shanghai in 2010 was 8.2533 million(SMSB, 2012). The variable t is the working time, which was 4h. The number of kitchen stoves in Shanghai is given as depicted in Figure 5(A). Calculated from equation (6), we determined the annual VOC emissions from cooking in Shanghai to be 7403.21±314.29t Yr^{-1}, as shown in Figure 5(B). The annual NMHC was found to be 11215.53±1074.36t Yr^{-1}.

Figure 5.

The third methodology based on EF_{hour} was calculated from equation (7):

$$S_{hour-type} = 365 \times \sum_i (EF_{hour} \times t \times Na) \tag{7}$$

where Na is the number of each cuisine type; t is the working time of the restaurant kitchens, 4h. The number of registered restaurants in Shanghai in 2012 was 36692 and can be divided into five categories: canteen/ super-huge/large types accounted for 7.4%; the percentage of medium and fast food restaurants was 18.0% and 5.0%, respectively; small scale and snack restaurants contributed to 60.0%; and the remaining 9.6% were tea houses and coffee bars. Using the information shown in Table 3, a value of 4124.33±120.47t Yr^{-1} was obtained for the annual total VOC emissions derived from cooking. The annual NMHC was found to be 6698.96±605.41t Yr^{-1}.

VOC Emission Inventories Based on Restaurant Scales. To estimate annual VOC emissions from restaurants in Shanghai based on restaurant scales, barbecue, fried food and family kitchens were not considered here, mainly because their operating modes are flexible, rendering them difficult for urban governance. Three methodologies associated with customers, kitchen stoves and cuisine types are given as equations (8)–(10), respectively.

$$S_{person-scale} = Q \times Nc \times EF_{person} \tag{8}$$

$$S_{kitchen\ stove-scale} = \sum N \times a \times t \times EF_{kitchen\ stove} \times 365 \tag{9}$$

$$S_{hour-scale} = \sum N \times t \times EF_{restruant} \times 365 \tag{10}$$

where Q is the Shanghai population; Nc is the customer dining frequency, and according to the aforementioned distribution of the percentage of the Shanghai population dining in restaurants per week, about an value of 100 times/year was obtained for Shanghai people eating in a restaurant(FDA, 2011). N is the number of restaurants for each scale; a is the number of kitchen stoves; t is the working time, 4h. Snacks and drinks/coffee/tea/ bars were classified as small scale restaurants. The emission factors shown in Table 2 were employed in the calculations. All parameters and the annual amount of VOC and NMHC emissions based on restaurant scales are listed in Table 3.

Table 3.

The calculated annual amount of VOC and NMHC emissions based on restaurant scales were less than those based on cuisine types for all three emission factors. One reason for this difference is the same as the interpretation given previously, that barbecue, fried food and family kitchens were not considered. Another reason for this difference is attributed to the lesser variances of EF among restaurants of the same scale.

Geographical Distribution of the Intensity of VOC and NMHC Emissions Produced by Cooking in Urban Shanghai.

According to the annual total VOC emissions calculated from restaurant scales, the geographical distribution of the intensities of VOC and NMHC emissions produced by cooking in Shanghai in 2012 are shown in Figure 6. Although Pudong and Minhang districts had the highest annual total VOC or NMHC emissions, the largest emission intensities appeared in Huangpu, Jing'an and Hongkou districts, which are located in urban centers – the emissions per unit area are larger than all other districts.

Figure 6.

Geographical Distribution of the annual total amount of VOC Emissions Produced by Cooking in China

Environmental Kuznets Curve(Dinda, 2004) indicates the economic capacity has a positive correlation with pollutant emissions prior to economy developed into a certain level, which presents an approximate linear relation. China is a developing country, which is located before the turning point in the curve. Therefore, according to the obtained yearly VOCs emissions of 100,000 people from catering business ($S_{hour\text{-}scale}$/Shanghai population * 100,000people), Shanghai catering consumption ability (as shown in Table SI4), and national catering consumption ability in China, the yearly VOCs emissions of 100,000 people in different provinces were obtained as Figure 7(a). It can be illustrated that VOCs emissions of 100,000 people from catering business in four municipalities are over 6t/year • 100,000people. Shanghai reached up to 8.16 t/year • 100,000people. Tianjin is the highest one among four municipalities, attaining to 11.23t/year • 10^5people. In addition, greater VOCs emissions of 100,000 people mainly occurred in provinces with high floating population and rich tourism resources. And furthermore, the yearly VOCs emissions of each province in China were obtained, as given by Figure 7(b). Shangdong and Gungdong provinces have the highest VOCs emissions, reaching up to 5680.53 t/year and 6122.43 t/year, respectively, nearly three times of Shanghai. The total annual VOCs emission is not only related to populations of different provinces, but also associated with local eating habits and economic conditions.

Figure 7.

Importance of Barbecue Emissions as a Source of Health Hazards. Considering the VOCs concentrations of barbeque emissions was the greatest in this study, and it is also the source nearest to the ground, hence its potential health effect are discussed. Acetaldehyde is classified as a group 2b carcinogen (possibly carcinogenic) by International Agency for Research on Cancer (IARC), with a limiting value of 0.003mg/m^3. But the acetaldehyde concentration emitted from barbeque was 0.34±0.07 mg/m^3 in this study. The monitored hexanal concentration was 0.26±0.02 mg/m^3, up to 8 times of the limiting value of 0.03 mg/m^3 set by German statutory accident insurance. Australian government and U.S Environmental Protection Agency (EPA) sets the limiting values of acrolein in workplaces as 0.23 and 0.24 mg/m^3, respectively. The monitored acrolein concentration was 0.24±0.04 mg/m^3 from barbeque emissions in this study.

CONCLUSIONS

This research sheds light on the significance of cuisine types and restaurant scales on VOC compositions, and their resulting chemical reactivities, that are entering into urban atmospheres from cooking emissions in Shanghai. Our results showed that alkane and oxygenated VOCs (O-VOCs) account for 13.26-65.85% and 1.67-50.30%, respectively to the VOC emissions produced by cooking. However, the VOCs with the largest OFP and SOAP were from the alkene (6.78-96.95%) and aromatic (73.75-98.86%) categories, respectively. Barbeque has the highest potential of hazardous health effect due to its significant higher emissions of acetaldehyde, hexanal, and acrolein.

The estimated annual total amount of VOCs is 4124.33-7818.04 t/year and 1355.11-2402.21 t/year based on S_{type} and S_{scale}, respectively. The VOCs emissions of 100,000 people from catering business are 8.16 t/year・100,000 people in Shanghai. According to the Environmental Kuznets Curve, the annual total amount of VOCs emissions from other provinces in China are obtained. Shangdong and Guangdong provinces reach up to 5680.53 t/year and 6122.43 t/year, respectively, which is not only related to populations of different provinces, but also associated with local cooking habits and economic conditions. Therefore, the annual amount of VOCs emission from catering industry in China is 66244.59 t/year, and 4.79 t/year・100,000 people.

Our quantitative analysis calls the attention of regulating authorities by providing them with the information needed to evaluate the major factors impacting on VOCs from cooking emissions in Shanghai as well as the whole nation. We suggest that large-and medium-scale restaurants should be regarded as the most important with respect to regulation of VOCs, and street barbeque should be taken seriously for its potential health hazard.

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Notes

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Figure Captions

Figure 1. Mass percentages of VOC species according to carbon numbers for each cuisine type

Figure 2. Comparison of compositions of VOCs emitted from different types of kitchens among different studies (A: Sichuan and Hunan cuisine; B: barbecue; C: family kitchen; D: fried food. SH: Shanghai-this study; BJ: Beijing-Zhang et al., 2011; HK: Hongkong-Yu Huang et al., 2011; MEX: Mexico- Mugica et al., 2000

Figure 3. Percentages of VOC categories contributing to OFP and the average MIR for each cuisine type

Figure 4. (A) Proportion and the number of people dining frequency for a week. (B) Proportion and the number of people eating in restaurants for each cuisines type. (C) Number of people eating in canteens and household kitchen, respectively. (D) VOCs emission of each cuisine type and the total annual VOCs emissions in Shanghai

Figure 5. (A) Number of each cuisine type and the corresponding number of kitchen stoves. (B) Annual total VOCs emissions of each type and the total VOCs emissions in Shanghai based on kitchen stove

Figure 6. Geographical distributions of the intensities of VOC and NMHC emission in Shanghai produced by cooking

Figure 7. (A) Geographical distributions of the yearly VOCs emissions of 100,000 people in different provinces. (B) Geographical distributions of the yearly VOCs emissions of each province in China

Table Captions

Table 1. Emission factors based on cuisine types

Table 2. Emission factors based on restaurant scales

Table 3. Parameters and emissions with respect to restaurants of various scales

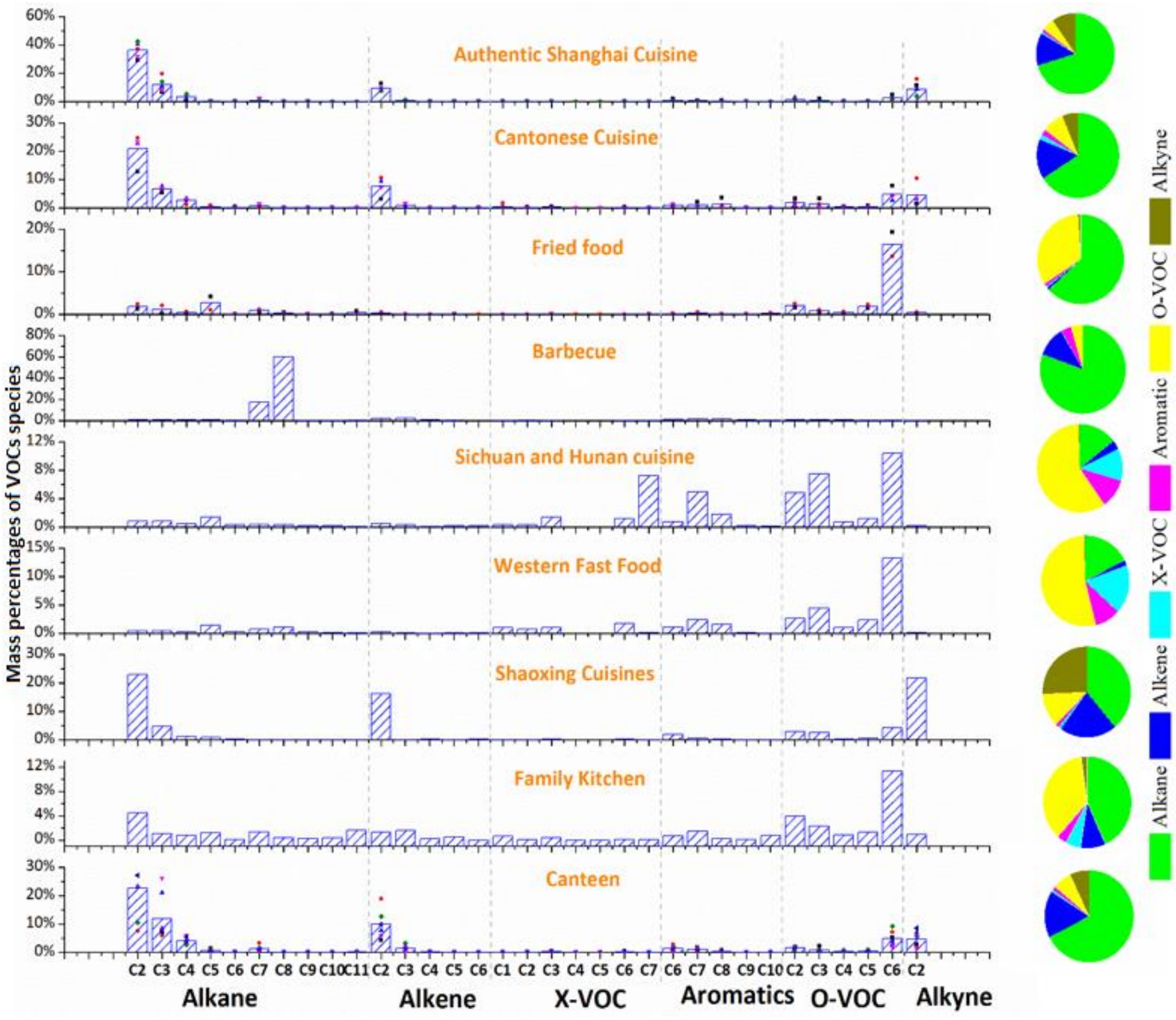

Authentic Shanghai Cuisine
Cantonese Cuisine
Fried food
Barbecue
Sichuan and Hunan cuisine
Western Fast Food
Shaoxing Cuisines
Family Kitchen
Canteen
Mass percentages of VOCs species
60%
40%
20%
0%
30%
20%
10%
0%
20%
10%
0%
80%
60%
40%
20%
0%
12%
8%
4%
0%
15%
10%
5%
0%
30%
20%
10%
0%
12%
8%
4%
0%
30%
20%
10%
0%
C2 C3 C4 C5 C6 C7 C8 C9 C10 C11 C2 C3 C4 C5 C6 C1 C2 C3 C4 C5 C6 C7 C6 C7 C8 C9 C10 C2 C3 C4 C5 C6 C2
Alkane
Alkene
X-VOC
Aromatics
O-VOC
Alkyne
Alkyne
O-VOC
Aromatic
X-VOC
Alkene
Alkane

Figure 1.

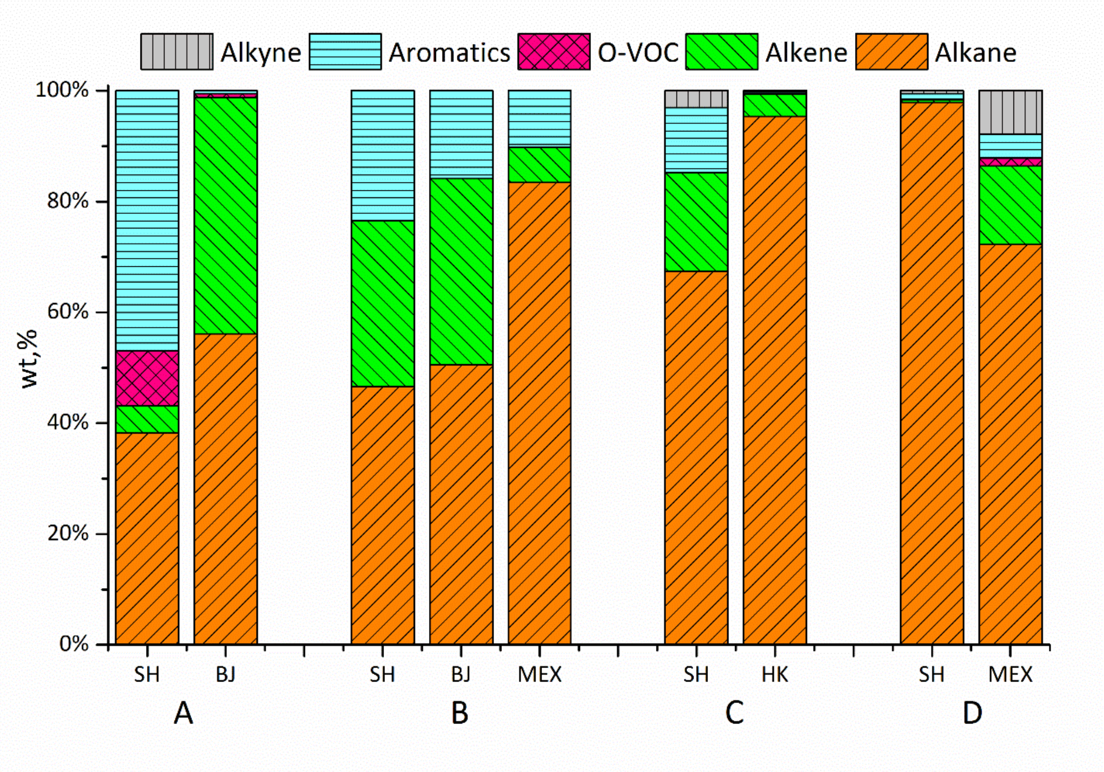

Alkyne
Aromatics
O-VOC
Alkene
Alkane
100%
80%
60%
40%
20%
0%
wt,%
SH
BJ
SH
BJ
MEX
SH
HK
SH
MEX
A
B
C
D

Figure 2.

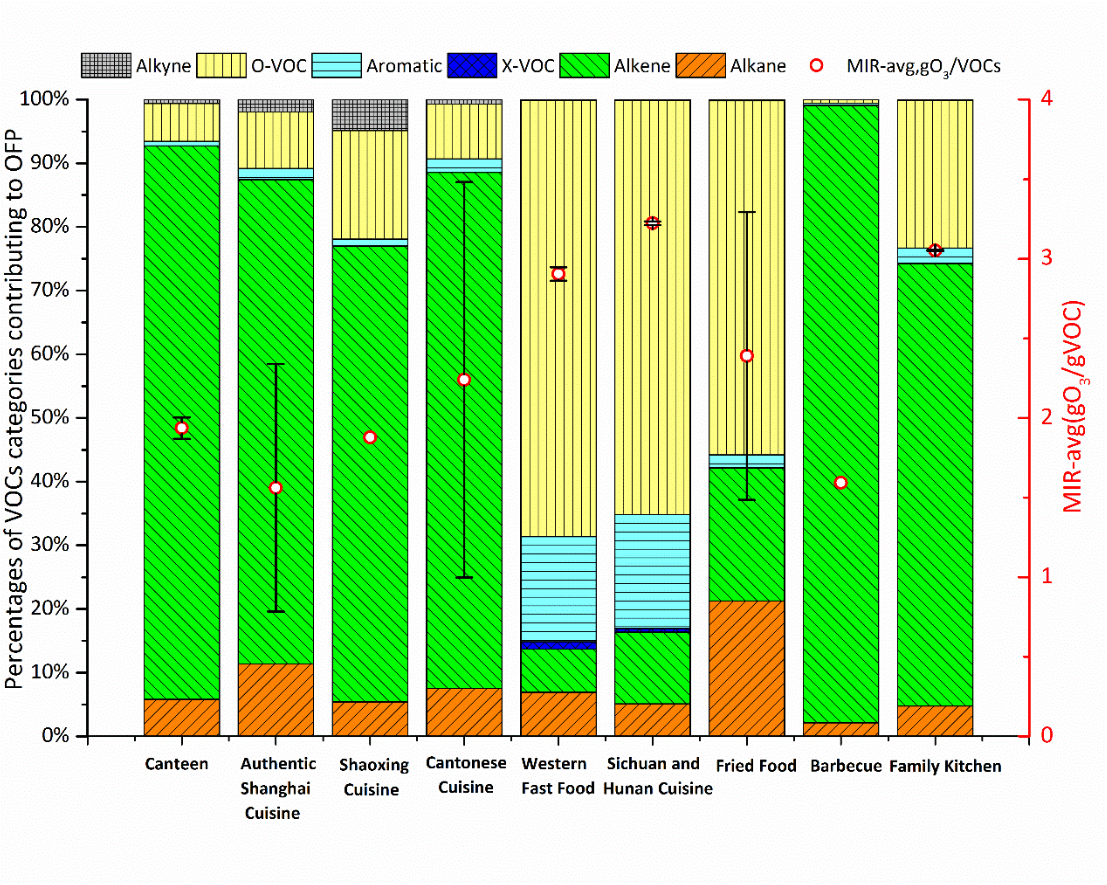

Alkyne
O-VOC
Aromatic
X-VOC
Alkene
Alkane
MIR-avg,gO$_3$/VOCs
Percentages of VOCs categories contributing to OFP
100%
90%
80%
70%
60%
50%
40%
30%
20%
10%
0%
MIR-avg(gO$_3$/gVOC)
4
3
2
1
0
Canteen
Authentic Shanghai Cuisine
Shaoxing Cuisine
Cantonese Cuisine
Western Fast Food
Sichuan and Hunan Cuisine
Fried Food
Barbecue
Family Kitchen

Figure 3.

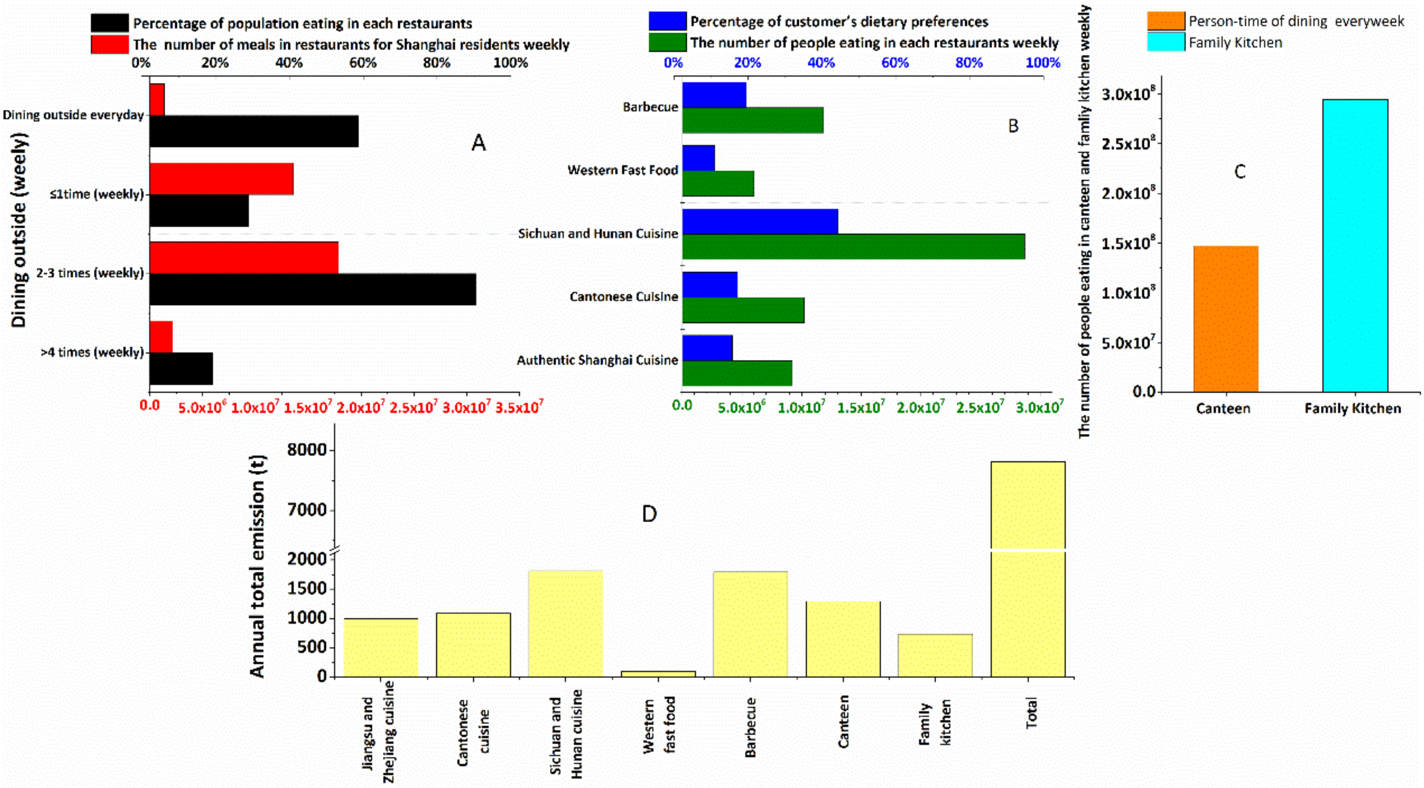

Percentage of population eating in each restaurants
The number of meals in restaurants for Shanghai residents weekly
0%
20%
40%
60%
80%
100%
Dining outside everyday
≤1time (weekly)
2-3 times (weekly)
>4 times (weekly)
Dining outside (weely)
A
0.0
5.0x10^6
1.0x10^7
1.5x10^7
2.0x10^7
2.5x10^7
3.0x10^7
3.5x10^7
Percentage of customer's dietary preferences
The number of people eating in each restaurants weekly
Barbecue
Western Fast Food
Sichuan and Hunan Cuisine
Cantonese Cuisine
Authentic Shanghai Cuisine
B
The number of people eating in canteen and family kitchen weekly
Person-time of dining everyweek
Family Kitchen
3.0x10^8
2.5x10^8
2.0x10^8
1.5x10^8
1.0x10^8
5.0x10^7
0.0
C
Canteen
Family Kitchen
Annual total emission (t)
8000
7000
2000
1500
1000
500
0
D
Jiangsu and Zhejiang cuisine
Cantonese cuisine
Sichuan and Hunan cuisine
Western fast food
Barbecue
Canteen
Family kitchen
Total

Figure 4.

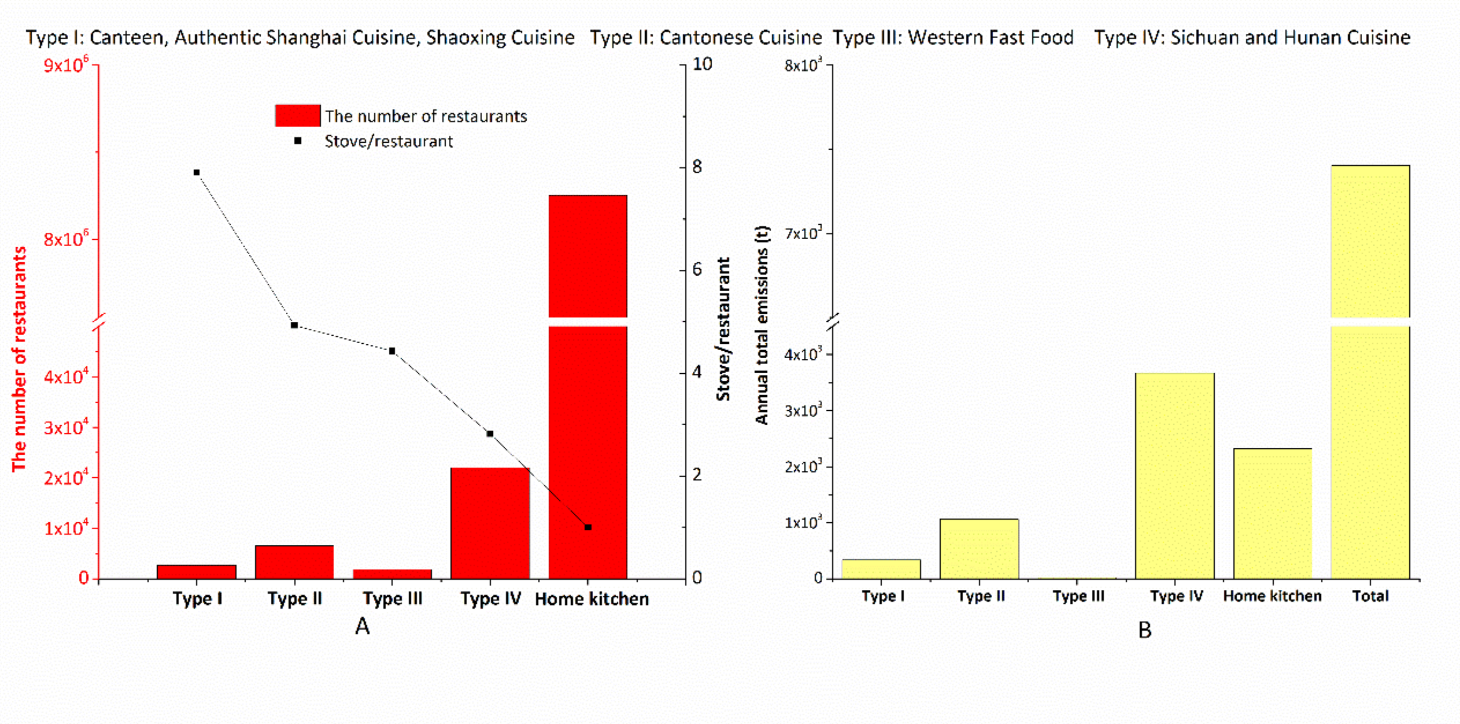

Type I: Canteen, Authentic Shanghai Cuisine, Shaoxing Cuisine
Type II: Cantonese Cuisine
Type III: Western Fast Food
Type IV: Sichuan and Hunan Cuisine
The number of restaurants
Stove/restaurant
The number of restaurants
9x10^6
8x10^6
4x10^4
3x10^4
2x10^4
1x10^4
0
Stove/restaurant
10
8
6
4
2
0
Type I
Type II
Type III
Type IV
Home kitchen
A
Annual total emissions (t)
8x10^3
7x10^3
4x10^3
3x10^3
2x10^3
1x10^3
0
Type I
Type II
Type III
Type IV
Home kitchen
Total
B

Figure 5.

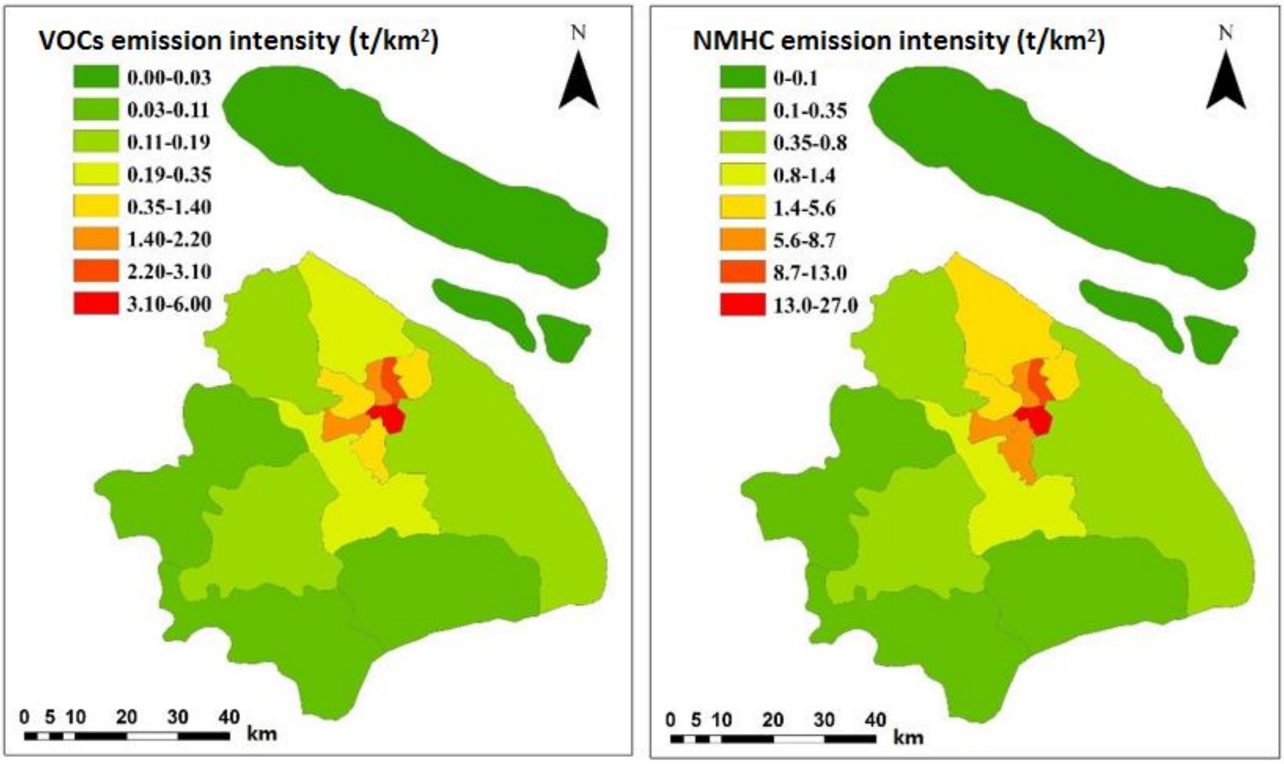
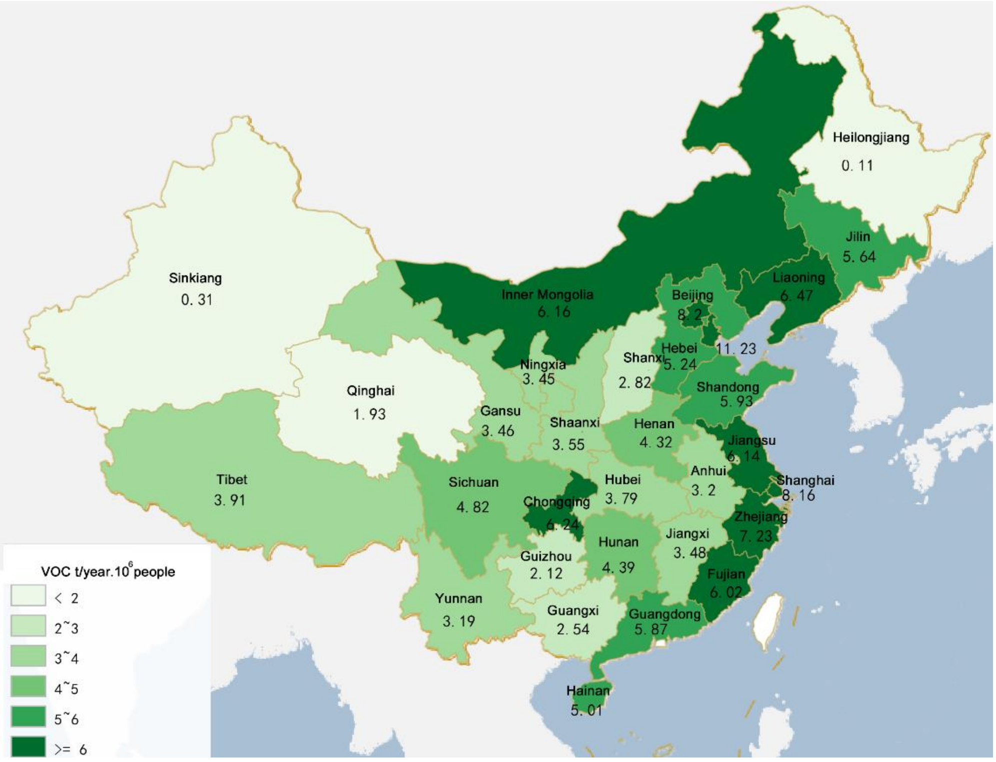

VOCs emission intensity (t/km2)
0.00-0.03
0.03-0.11
0.11-0.19
0.19-0.35
0.35-1.40
1.40-2.20
2.20-3.10
3.10-6.00
N
0 5 10 20 30 40
km
NMHC emission intensity (t/km2)
0-0.1
0.1-0.35
0.35-0.8
0.8-1.4
1.4-5.6
5.6-8.7
8.7-13.0
13.0-27.0
N
0 5 10 20 30 40
km

Figure 6.


Heilongjiang
0. 11
Jilin
5. 64
Liaoning
6. 47
Inner Mongolia
6. 16
Beijing
8. 2
11. 23
Hebei
5. 24
Shanxi
2. 82
Shandong
5. 93
Sinkiang
0. 31
Ningxia
3. 45
Qinghai
1. 93
Gansu
3. 46
Shaanxi
3. 55
Henan
4. 32
Jiangsu
6. 14
Tibet
3. 91
Sichuan
4. 82
Chongqing
6. 24
Hubei
3. 79
Anhui
3. 2
Shanghai
8. 16
Zhejiang
7. 23
Jiangxi
3. 48
Hunan
4. 39
Guizhou
2. 12
Fujian
6. 02
Yunnan
3. 19
Guangxi
2. 54
Guangdong
5. 87
Hainan
5. 01
VOC t/year.10^6 people
< 2
2~3
3~4
4~5
5~6
>= 6

(a)

(b)

Figure 7

Table 1

	$EF_{people-type}$ (g/person)		**$EF_{kitchen\ stove-type}$ (g/h·stove)**		**$EF_{hour-type}$ (g/h)**	
Cuisine (Number of samples)	**VOCs**	**NMHC (by carbon)**	**VOCs**	**NMHC (by carbon)**	**VOCs**	**NMHC (by carbon)**
Canteen (27)	0.01±0.00	0.10±0.03	1.97±1.33	16.18±10.96	15.76±5.94	129.40±0.033
Authentic Shanghai Cuisine(6)	2.54±1.30	15.55±7.96	9.09±2.49	55.54±15.22	111.04±30.43	634.56±7.96
Shaoxing Cuisine(2)	2.26±0.00	13.22±0.00	12.52±0.00	61.33±0.00	225.59±0.00	1030.22±0.00
Cantonese Cuisine(8)	1.96±1.24	8.41±5.30	12.04±7.14	55.46±32.89	78.41±38.66	358.54±176.77
Western Fast Food(2)	0.32±0.04	0.60±0.08	1.86±0.24	3.47±0.48	11.15±1.44	20.84±2.69
Sichuan and Hunan Cuisine(4)	0.17±0.00	0.25±0.00	5.94±0.03	8.18±0.04	17.80±0.09	24.53±0.13

Table 2

	$EF_{people-scale}$ (g/person)		**$EF_{kitchen\ stove-scale}$ (g/h·stove)**		**$EF_{hour-scale}$ (g/h)**	
Scale (Number of samples)	**VOCs**	**NMHC (by carbon)**	**VOCs**	**NMHC (by carbon)**	**VOCs**	**NMHC (by carbon)**
Canteen (27)	0.01±0.00	0.1±0.032	1.97±1.33	16.18±10.96	15.76±5.94	129.4±48.80
Extra-large (4)	1.77±0.32	5.72±1.02	8.57±1.49	40.84±7.11	128.94±22.88	285.85±50.71
Large (6)	3.81±0.76	19.67±3.95	13.56±2.73	70.23±14.114	189.78±38.14	983.26±197.61
Medium (6)	1.97±0.26	8.41±1.10	12.03±3.53	55.46±16.25	78.41±22.98	358.53±105.06
Small (4)	0.18±0.00	0.25±0.00	5.94±0.03	8.18±0.04	17.82±0.09	24.53±0.13
Fast food (2)	0.32±0.04	0.60±0.08	1.86±0.24	3.47±0.45	11.15±1.44	20.84±2.69

Scales	N	a	$S_{kitchen-stove-scale}$ (t/year)		$S_{hour-scale}$ (t/year)		$S_{people-scale}$(t/year)	
			VOCs	NMHC	VOCs	NMHC	VOCs	NMHC
Canteen	208	2.93	1.77±0.12	14.44±4.22	4.80±1.23	39.40±4.56	-	-
Extra large	100	22.25	27.92±3.24	133.03±34.52	18.89±2.33	41.86±6.73	-	-
Large	2392	8.54	405.53±24.57	2100.31±134.56	664.60±56.34	3443.25±456.22	-	-
Medium	6590	4.93	572.19±33.11	2637.88±245.67	756.52±45.67	3459.04±243.20	-	-
Small	7842	2.97	202.54±12.59	278.92±4.56	204.57±19.79	281.59±15.34	-	-
Fast food	1843	4.43	22.23±5.13	41.49±2.47	30.08±4.56	56.22±7.54	-	-

Snacks	14183	2.69	103.50±7.08	193.10±34.23	231.50±12.58	432.64±45.80	-	-
Drinks/Coffee/Tea/Bar	3534	2.02	19.44±2.33	36.27±3.56	57.69±6.98	107.80±7.57	-	-
Total	**36692**	**-**	**1355.11±107.24**	**5435.42±185.45**	**1968.61±98.57**	**7861.788±267.56**	**2402.21±145.67**	**10396.77±345.79**

Table 3