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A comprehensive study of volatile organic compounds from the actual emission of Chinese cooking

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Abstract

China's current perspective for big cities were filled with great population, great number of restaurants, growing gaseous pollutants and great pollution. Volatile organic compounds (VOCs) were main components of catering industry gaseous pollutants. In this study, we collected and analyzed VOCs from Home cooking (HC), Sichuan and Hunan cuisine (S&H), Shandong cuisine (SD), Anhui cuisine (AH), Beijing cuisine (BJ) and Barbecue (BBQ). The result showed that BBQ gave the highest VOCs concentration ($6287.61 \mu\text{g}\cdot\text{m}^{-3}$), followed by HC ($1806.11 \pm 2401.85 \mu\text{g}\cdot\text{m}^{-3}$), SD ($2238.55 \pm 2413.53 \mu\text{g}\cdot\text{m}^{-3}$), AH ($1745.89 \mu\text{g}\cdot\text{m}^{-3}$), S&H ($1373.58 \pm 1457.45 \mu\text{g}\cdot\text{m}^{-3}$) and BJ ($288.81 \mu\text{g}\cdot\text{m}^{-3}$). The abundance of alkane was higher among BBQ, Anhui Cuisine and HC with the proportion from 33% to 71%. SD contained higher halohydrocarbons proportion (33%). BJ characterized by high-oxygenated volatile organic compounds proportion (50%). The ozone formation potential (OFP) of BBQ was much greater than other cuisines. The degree of stench pollution from cooking VOCs of HC was 17.51 ± 16.95 , followed by S&H (15.77 ± 16.85), SD (15.12 ± 14.17), AH (16.29), BBQ (2.58) and BJ (1.81). Benzene had highest life cancer risk (LCR) in SD ($2.11 \times 10^{-5} \pm 3.12 \times 10^{-5}$), following HC ($4.50 \times 10^{-6} \pm 3.83 \times 10^{-6}$)

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and S&H ($4.08 \times 10^{-6} \pm 4.49 \times 10^{-6}$). Acrolein had a high hazard index (*HI*) in HC (789.81 ± 768.77), following AH (728.78), S&H (689.89 ± 776.07), SD (664.29 ± 648.77), BBQ (65.93) and BJ (62.84).

Keywords: volatile organic compounds, cooking cuisines, ozone formation potential, stench pollution, health risk assessment

1. Introduction

Due to rapid economic development, consumers spend more money in catering industry, especially who live in megacity. According to the data from the National Bureau of Statistic of China, up to December 2019, the total value of retail sales of restaurants in China was 4.7 trillion RMB. Behind the blooming of catering industry, gaseous pollutants were increasing considerably (Barletta et al. 2008; Jiamin et al. 2015; Chiungyu et al. 2017; Lina et al. 2017). Researches showed that catering emission took great affection to the resident environment. Catering emission mainly consisted of oil smoke, particular matter (Lingyan et al. 2004; Lina et al. 2018) and volatile organic compounds (Chen et al. 2018; Shaoqing 2017). VOCs could generate secondary particulate via complex chemical reactions and the reaction products could suspension in the air for a long time (Dall'Osto et al. 2015). VOCs threaten atmosphere environment and resident health (Wang et al. 2018; Kabir et al. 2011; Lina et al. 2018). Some VOCs showed great ozone formation potential (OFP) (Carter 2010; Shuiyuan et al. 2016; Zhiyuan et al. 2017; Hongli et al. 2018). Additionally, the stench of VOCs had a negative effect on the ambient environment (Lina et al. 2017; Kabir et al. 2011; Xufeng et al. 2019; Xiuyan et al. 2012; Yoshio 2003).

Studies of catering industry VOCs mainly focused on the total average concentration and characteristics of components. Wang et al. (2018) analyzed 99 VOCs from C₁ to C₇, obtained the emission factors based on per person, per kitchen stove and per hour, established VOC emission inventories based on restaurant scales cuisine types ($4124.33 \sim 7818.04 \text{ t} \cdot \text{y}^{-1}$) and restaurant scales ($1355.11 \sim 2402.21 \text{ t} \cdot \text{y}^{-1}$) and gave the conclusion that medium-scaled restaurants should be regarded as the most important with

respect to regulation of VOCs. The research results provided a methodology for the determination of emission factors and the establishment of emission inventories, and aroused managers to pay more attention to catering emissions. Yi et al. (2019) found the benzene series emission for three edible oils were high and give a conclusion that the total VOCs emission of catering industry could be a function of edible oils consumption. However, the study focused on the emissions of edible oils, the real emissions of cooking process were neglected. Cheng et al. (2016) measured 51 VOCs including 26 alkanes, 9 alkenes and 16 aromatics and compared the characteristics among Home Cooking, Shandong cuisine, Hunan Cuisine and Barbecue and compared the ozone formation potential (OFP) and stench index. In Cheng et al. (2016) research, coefficient of divergence method was used to analyze the level of VOCs similarity, and great difference was found between chemical profiles of different cuisine types. Hongli et al. (2018) compared the characteristics among Authentic Shanghai cuisine, Cantonese cuisine, fired food, street barbecue, Sichuan & Hunan cuisine, Western fast food, Shaoxing cuisine, Canteen and Home cooking in Shanghai and alerted that street Barbecue had potential health hazard with high concentration of acetaldehyde. Min et al. (2017) gave the conclusion that C₁ to C₃ VOCs whose main components were acetaldehyde, formaldehyde, and acrolein, took 72.27% of VOCs emission of traditional Beijing roast duck. To obtain the real emission character of Beijing roast duck, the sampling site which Min et al. (2017) chose was before the purification device. Zhiyuan et al. (2017) measured carbonyl compounds emission intensity for different cooking styles and found barbecue > frying > teppanyaki > stir-frying, and for food materials they found meat dishes > non-meat dishes. Chunyang et al. (2011) analyzed the characterization of VOCs emitted from 5 restaurants and found the proportion of alkanes, alkenes and aromatics were similar and the proportion of halohydrocarbons existed great difference. However, the sampling sites in their research (Zhiyuan et al. 2017; Chunyang et al. 2011) were after the purification

devices, which may not reflect the real emission characterization of VOCs cooking emission.

VOCs were one of the main sources of ozone, especially aromatics and alkene (Carter 1994, 2010). Yafei et al. (2020) measured 99 VOCs in Beijing during ozone pollution days and found the combustion source generated most VOCs. Vehicle exhaust contributed the most (34.52%) of the total OFP in Shenyang (Zhuobiao et al. 2019). For commercial cooking, Qiusheng et al. (2015) founded the average maximum reactivity coefficient (MIR) of Chinese restaurant ($3.34 \text{ gO}_3 \cdot \text{gVOCs}^{-1}$) and barbecue ($2.93 \text{ gO}_3 \cdot \text{gVOCs}^{-1}$) were at the medium level among different emission sources. Shuiyuan et al. (2016) calculate the average MIR of Shandong cuisine, Hunan cuisine and Barbecue were 4.01, 4.31, 4.05 and $4.81 \text{ gO}_3 \cdot \text{gVOCs}^{-1}$, respectively. The average MIR did not show great difference between cuisine types.

The VOCs emitted from catering industry had considerable stench (Buonanno et al. 2009), few studies concentrated on it. According to Cheng (Shuiyuan et al. 2016), only propyl benzene and 1,4-diethylbenzene emitted from barbecue existed a certain degree of stench pollution. Kabir's (Kabir et al. 2011) conclusion strongly suggested that the odor emission was depended on the gradients and cooking process.

To have a more comprehensive knowledge of the catering industry VOCs components and its affect to ambient air quality, 27 restaurants in Beijing were selected and classified as Home cooking (HC), Sichuan and Hunan cuisine (S&H), SD, Anhui cuisine (AH), Beijing cuisine (BJ) and BBQ. 101 VOCs from C_3 to C_{12} were detected by gas chromatography-mass spectrometry. The objection of this study mainly include: 1) analyzing the VOCs emission characteristics from different cooking styles; 2) calculating the OFP of different cooking styles; 3) investigating the stench of index of VOCs.

2. Materials and Method

2.1 On-site sampling collection

From May 1st to May 10th in 2019, 27 VOCs samples from 27 restaurants, which including 6 cuisine

types, were collected during the peak cooking periods (10:30~13:00 and 17:00~20:00) of the restaurants in Beijing. The cuisine types included S&H (8 restaurants), HC (12 restaurants), SD (4 restaurants), AH (1 restaurant), BBQ (1 restaurant) and BJ (1 restaurant).

The emissions of VOCs from restaurants exhausted through exhaust pipes. The airflow was unstable near elbows and junctions and that would decrease the accuracy of the data. To obtain a stable airflow, the sampling points and sampling method were selected via Determination of practices and sampling method of gaseous pollutants emitted from exhaust gas of stationary source (GB/T 16157-1996).

The sampling system included the following sections: 1) Sampling pipe was a section of Pitot tube, which was linked to the sampling point. 2) Fume filter could remove the oil fume and particulate matter in the sample gas. 3) Vacuum box (Restek) was used to transport the emissions into the Tedlar bag (10L). 4) Aerometer was used to measure the air flowrate of the exhaust gas and transfer the results under standard temperature and pressure.

Samples were collected, with a stable $0.5 \text{ L}\cdot\text{min}^{-1}$ flow rate and persisting 30 minutes, right before the purification device during the peak cooking periods.

2.2 Sample analysis

The VOCs of cooking oil were first analyzed by gas chromatography-mass spectrometry (GC-MS) in 1995 (Overton et al. 1995). The invention of purge & trap decreased the method detection limit (MDL) of the target VOC. In this study, 101 VOCs containing alkanes, alkenes, oxygenated volatile organic compounds (OVOCs), carbon disulfide, aromatics and halohydrocarbons were analyzed by GC-MS. The collected samples were pumped into the pre-concentrator which include 3 modules. Module 1 was set to remove H_2O and CO_2 and O_2 at -150°C by liquid nitrogen, and the samples were recovered by desorbing at 10°C to remove most of the liquid H_2O . Samples were cooled to -30°C in Module 2 to enrich the

concentration of VOCs, and then the samples were heated to 50°C to release the VOCs into Module 3. Samples were cooled to -150°C by liquid nitrogen in Module 3, and reheat to 80°C to make the VOCs samples vaporized rapidly. Then the concentrated VOCs samples were carried into GC-MS. A DB-624 column (60 m × 250 μm × 1.4 μm) was used in GC. The column was set at an oven temperature of 35°C for 5 minutes, then raised to 160 °C at a rate of 5 °C•min⁻¹, and finally raised to 220°C at a rate of 20 °C•min⁻¹.

Tedlar bags were pre-cleaned and vacuumed by nitrogen (N₂, 99.99%) to remove any organic compounds before sampling. The Pitot tube was purged with the target gas before the sampling procedure. TO-15 (65 components, 1ppm, Linde), PAMs (57 components, 1ppm, Linde) and Internal Standard Gas (4 components, 1ppm, Linde) were used to generated the calibration curve. The detailed information of 101 VOCs was listed in **table 1**. The calibration curves for 101 target VOCs were performed under different concentrations and the correlation coefficients were all above 0.99. The MDL of 101 species were all ranged from 0.07~0.63 μg•m⁻³. The concentration of 101 VOCs species were obtained from the calibration curves.

Table 1 Compounds information

Serial	Compound	CAS	MIR*	Serial	Compound	CAS	MIR*
1	propane	74-98-6	0.49	52	methyl isobutyl ketone	43209-75-2	3.88
2	iso-butane	75-28-5	1.23	53	tetrahydrofuran	109-99-9	2.62
3	n-butane	106-97-8	1.15	54	diethylene dioxide	123-91-1	2.62
4	iso-pentane	78-78-4	0.37	55	2-hexanone	591-78-6	3.14
5	n-pentane	109-66-0	1.31	56	benzene	64268-28-6	0.72
6	2,2-dimethyl butane	75-83-2	1.17	57	toluene	108-88-3	4.00
7	2,3-dimethylbutane	79-29-8	0.97	58	ethylbenzene	100-41-4	3.04
8	2-methylpentane	107-83-5	1.50	59	o-xylene	95-47-6	7.64
9	cyclopentane	287-92-3	1.96	60	isopropyl benzene	68936-98-1	2.52
10	3-methyl pentane	96-14-0	1.80	61	propylbenzene	103-65-1	2.03
11	n-hexane	110-54-3	1.24	62	3-ethyltoluene	620-14-4	7.39
12	2,4-dimethylpentane	108-08-7	--	63	4-ethyltoluene	25550-14-5	4.44
13	methylcyclopentane	96-37-7	1.70	64	1,3,5-trimethylbenzene	108-67-8	11.76
14	2-methylhexane	591-76-4	--	65	2-ethyltoluene	611-14-3	5.59

15	cyclohexane	110-82-7	1.25	66	1,2,4-trimethylbenzene	95-63-6	8.87
16	2,3-dimethylpentane	565-59-3	1.34	67	1,4-dichlorobenzene	106-46-7	--
17	3-methylhexane	589-34-4	1.61	68	1,2,3-trimethylbenzene	526-73-8	11.97
18	2,2,4-trimethylpentane	540-84-1	1.26	69	1,3-diethylbenzene	141-93-5	7.10
19	n-heptane	142-82-5	1.07	70	1,4-diethylbenzene	105-05-5	4.43
20	methylcyclohexane	108-87-2	1.70	71	naphthalene	91-20-3	3.30
21	2,3,4-trimethylpentane	565-75-3	--	72	dichlorodifluoromethane	75-71-8	--
22	2-methylheptane	592-27-8	1.07	73	dichlorotetrafluoroethane	1320-37-2	--
23	3-methylheptane	589-81-1	1.24	74	chloromethane	74-87-3	--
24	octane	111-65-9	0.90	75	methyl bromide	74-83-9	--
25	dibromo-monochloro-methane	4371-77-1	--	76	chloroethane	75-00-3	--
26	1,2-dibromoethane	106-93-4	--	77	trichlorofluoromethane	75-69-4	--
27	nonane	111-84-2	0.78	78	trichlorotrifluoroethane	26523-64-8	--
28	decane	124-18-5	0.68	79	dichloromethane	75-09-2	--
29	undecane	1120-21-4	0.61	80	1,1-dichloroethane	75-34-3	--
30	dodecane	112-40-3	0.55	81	trichloromethane	8013-54-5	--
31	propene	115-07-1	11.66	82	1,1,1-trichloroethane	71-55-6	--
32	1-butene	106-98-9	9.73	83	1,2-dichloroethane	107-06-2	--
33	1,3-butadiene	25339-57-5	12.61	84	1,2-dichloropropane	78-87-5	--
34	2-butene	104-01-7	16.16	85	bromodichloromethane	75-27-4	--
35	cis-2-butene	590-18-1	14.24	86	1,1,2-trichloroethane	25323-89-1	--
36	1-pentene	25377-72-4	7.21	87	bromoform	2909-52-6	--
37	trans-2-pentene	109-68-2	10.56	88	1,1,2,2-tetrachloroethane	79-34-5	--
38	isoprene	78-79-5	10.61	89	vinyl chloride	93050-82-9	--
39	cis-2-pentene	627-20-3	10.38	90	vinylidene chloride	75-35-4	--
40	1-hexene	592-41-6	5.49	91	(E)-1,2-Dichloroethene	156-60-5	--
41	styrene	100-42-5	1.73	92	cis-1,2-dichloroethene	156-59-2	--
42	hexachlorobutadiene	87-68-3	--	93	trichloroethene	79-01-6	--
43	carbon disulfide	75-15-0	--	94	trans-1,3-dichloropropene	10061-02-6	--
44	acrolein	107-02-8	7.45	95	cis-1,3-dichloropropene	10061-01-5	--
45	methyl methacrylate	80-62-6	15.61	96	tetrachloroethylene	127-18-4	--
46	methyl tertiary-butyl ether	1634-04-4	0.73	97	chlorobenzene	108-90-7	--
47	vinyl acetate	108-05-4	3.20	98	1,3-dichlorobenzene	541-73-1	--
48	acetone	68937-52-0	0.36	99	benzyl chloride	100-44-7	--
49	isopropanol	67-63-0	--	100	1,2-dichlorobenzene	95-50-1	--
50	2-butanone	78-93-3	1.48	101	1,2,4-trichlorobenzene	120-82-1	--
51	ethyl acetate	141233-21-8	0.63				

--No data available; * MIR represented maximum reactivity coefficient (Carter 2010)

3 Data calculation

3.1 Normalized concentration calculation

The concentrations of 101 VOCs were normalized by equation (1).

$$C_{std} = C_{test} \times \frac{Q_{test}}{nQ_{std}} \quad (1)$$

Where C_{std} and C_{test} represent the normalized and the test VOC concentration ($\mu\text{g}\cdot\text{m}^{-3}$). n represents the number of the normalized stoves. Q_{test} represent the result of aerometer ($\text{m}^3\cdot\text{h}^{-1}$) and Q_{std} was equal to $2000 \text{ m}^3\cdot\text{h}^{-1}$.

3.2 Ozone formation potential Calculation

Ozone formation was related to VOCs emission (Hairong et al. 2010). Previous studies mainly focused on other sources, such as gasoline-related emissions, petrochemicals, liquefied petroleum gas, et al. (Yu et al. 2007; Rongrong and Shaodong 2017; Qiusheng et al. 2015; Zhuobiao et al. 2019; Garzón et al. 2015; Gang et al. 2016; Jeeranut et al. 2012). In this study, 101 VOC species were detected. Previous studies calculated the ozone formation potential (OFP) for different cuisine types (Hongli et al. 2018; Shuiyuan et al. 2016). The contribution of individual VOC to OFP was evaluated by the MIR which was first claimed by Carter (Carter 1994, 2010). The MIR calculating based on the following equation:

$$OFP_i = VOC_{S_i} \times MIR_i \quad (2)$$

Where OFP_i was defined as the ozone formation potential for individual VOC_i . MIR_i was the maximum reactivity coefficient of compound i , and the detailed data of MIR was based on Carter's research (Carter 2010).

3.3 Stench calculation

The odor of catering VOCs affected the ambient air quality and threatened the resident's health (Kabir et al. 2011). Shuiyuan et al (2016) evaluated the stench index by the following equation:

$$M_i = \frac{C_i}{\mu_i} \quad (3)$$

Where M_i represented the diluted multiples of component i , C_i and μ_i represented the mass concentration ($\mu\text{g}\cdot\text{m}^{-3}$) and odor thresholds ($\mu\text{g}\cdot\text{m}^{-3}$) of component i (Yoshio 2003), respectively.

3.4 Health risk assessment

Health risk assessment of cooking VOCs mainly focused on indoor pollutants (Chunyan et al. 2017). There was still lack of reports on health risk assessment about cooking VOCs in emission gas. In our research, we assumed that the emitted gas was directly inhaled by the residents. The exposure concentrations (EC) of were estimated following inhalation assessment via equation (China. 2019; USEPA. 1992) (4).

$$EC = \frac{CA \times ET \times EF \times ED \times IR}{AT} \quad (4)$$

Where CA represented the VOCs concentration ($\mu\text{g}\cdot\text{m}^{-3}$), ET represented exposure time and the value was $4 \text{ h}\cdot\text{d}^{-1}$ according to *Technical specifications for health risk assessment of ambient air pollution* (China 2019), EF represented exposure frequency ($\text{d}\cdot\text{y}^{-1}$), ED represented exposure duration (y), IR represented inhalation rate ($\text{m}^3\cdot\text{h}^{-1}$), AT represented average lifetime (h).

Non-cancerous risk was calculated by hazard quotient (HQ). HQ for pollutants was calculated as dividing the EC by the pollutant specific reference concentration (RfC) (USEPA-IRIS 2009; China 2019).

$$HQ = \frac{EC}{RfC \times 1000 \mu\text{g}/\text{m}\text{g}} \quad (5)$$

Collective non-cancerous effects due exposure to several pollutants were known as the hazard index (HI). The acceptable value of HI was 1 (China 2019).

$$HI = \sum HQ \quad (6)$$

Lifetime cancer risk (LCR) was calculated by equation (7) The acceptable value of LCR was 1.00×10^{-6} (China 2019).

$$LCR = EC \times IUR \quad (7)$$

Where IUR represented the inhalation unit risk which data can be found in USEPA website (USEPA-IRIS 2009).

4. Result and Discussion

4.1 VOCs characterizes of different cooking cuisines

101 VOCs species were measured including 28 alkanes, 10 alkenes, 12 OVOCs, carbon disulfide, 16 aromatics and 34 halohydrocarbons. The average concentrations of detected compounds emitted from different cooking styles were demonstrated in Table 1 and Fig 2. The total VOCs (TVOCs) of S&H, HC, SD, AH, BBQ and BJ were 1373.58 ± 1457.45 , 1806.11 ± 2401.85 , 2238.55 ± 2413.53 , 1745.89 , 6287.61 and $288.81 \mu\text{g}\cdot\text{m}^{-3}$. TVOCs concentration from BBQ was the highest. That might due to spices, meat and charcoal, which could generate more VOCs during the cooking process. The results were consistent with previous study that Barbecue gave the highest VOCs concentrations among all the cooking styles from $3494 \mu\text{g}\cdot\text{m}^{-3}$ to $12470.6 \mu\text{g}\cdot\text{m}^{-3}$, followed by Hunan cuisine, Home cooking and et al. (Shuiyuan et al. 2016; He et al. 2020). However, different concentration levels were exhibited for the same cuisine form. The reason may be attributed to Non-normalized concentration, different species being tested and different sampling time. VOCs concentration in the exhaust would be impacted largely by the volume of air drawn through the collection hood over the stove. While the emission of cooking fume was intermittent. Honghong et al. (2019) focused on the emission of NMHC and benzene series of three commonly used cooking oils. The results exhibited that the emissions were increased with the rise of heating temperature, especially when the temperature was over $200\text{ }^{\circ}\text{C}$. But cooking fume was generated by edible oil and food after a series of complex chemical reaction at a high temperature, the emissions of oils does not reflect the actual emissions.

The distribution of VOCs categories in different cuisines were show in **fig.1**. *1)* Alkanes contributed 71% of VOCs emission in BBQ, which was due to charcoal burning (Bilsback et al. 2019) and incomplete combustion of fats derived from meat (Umano et al. 1987). The main alkane compounds of BBQ were

propane ($605.37 \mu\text{g}\cdot\text{m}^{-3}$), iso-butane ($1689.77 \mu\text{g}\cdot\text{m}^{-3}$) and n-butane ($2091.62 \mu\text{g}\cdot\text{m}^{-3}$). For HC, S & H, SD, AH and BJ, the proportion of alkane were similar and in a high level (16%~42%). Because the cooking methods of the above cuisines were similar, which mainly stir-fry. The alkanes were coming from fugitive emissions form natural gas and liquid petroleum gas and from the decomposition of cooking oil. 2) Alkenes contributed 28% of VOCs emission in BBQ. The main alkane compounds of BBQ were 1-butene ($651.93 \mu\text{g}\cdot\text{m}^{-3}$) and 2-butene ($1059.96 \mu\text{g}\cdot\text{m}^{-3}$), which may share the same original sources with iso-butane and n-butane because the structure, molecular weight and concentrations of these compounds were similar. For HC, S&H, SD, AH and BJ, the proportions of alkane were similar and in a low level (4%~13%).3) OVOC contributed 50% of VOCs emission in BJ, characterized by high acetone proportion (80% of OVOC). The main OVOC compounds of S&H, AH, SD, HC and BBQ were acrolein, acetone and ethyl acetate. Acrolein may came from heating cooking oils and fat (Umano et al. 1987). 4) Carbon disulfide ranged from $34.49 \mu\text{g}\cdot\text{m}^{-3}$ to $73.15 \mu\text{g}\cdot\text{m}^{-3}$ for HC, S&H, SD, AH and BJ and BBQ had a low concentration ($1.82 \mu\text{g}\cdot\text{m}^{-3}$). The results did not had great affect towards TVOCs.5) Aromatics contributed 11%~14% in S&H, AH, SD, HC and BJ and took a minimal ratio in BBQ. 6) Halohydrocarbons contributed 33% of VOCs emission in SD, that maybe the result of the seasoning which contained large amount of sodium chloride were widely used in SD.

Notably, all the research results showed that alkanes are the dominant species. For example, Cheng et al. (2016) obtained 26 alkanes in Home cooking, Shandong cuisine, Hunan cuisine and Barbecue with the abundance were 63.8%, 59.4%, 61.3% and 34.7%, respectively. He et al (2020) obtained 8 alkanes in Sichuan and Hunan cuisine and Family cuisine with the abundances were 31.8% and 47.9%, respectively. Comprehensively, the differete abundance of VOCs among all the cooking fumes maybe attribute to differences in the ingredients, operating temperatures and operation modes.

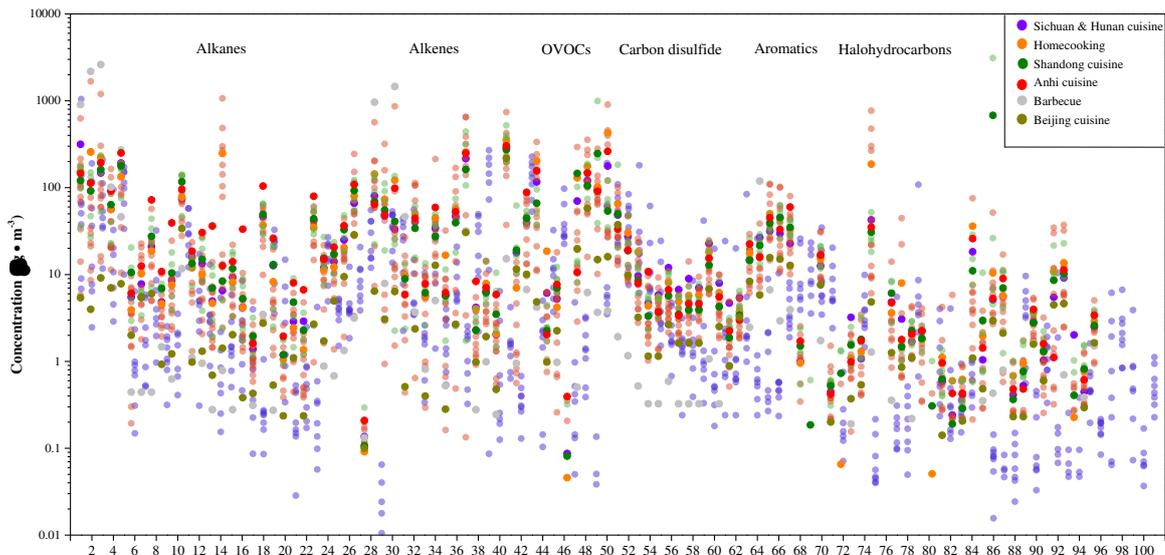


Fig.1. VOCs characterizes of different cooking cuisines (The abscissa axis was the serial numbers of compounds which were listed in **table 1**)

4.2 Ozone formation potential of different cooking cuisines

In present, more attention was paid to the contribution of VOCs from industrial sources to ozone generation, such as petrochemical industry, chemical industry and furniture painting. However, catering industry should be taken more attention in the case of fewer industrial sources in urban area. In this study, 64 VOCs species in the 101 detected VOCs species were selected to calculate the OFP of each individual VOC. The rank of total OFP value was not similar with the rank of TVOCs. The OFP for different cuisine types were listed in **table 2 and fig 2**. BBQ had the highest total OFP of VOCs ($28927.19 \mu\text{g}\cdot\text{m}^{-3}$), following AH ($5188.10 \mu\text{g}\cdot\text{m}^{-3}$), SD ($4710.10 \pm 3528.30 \mu\text{g}\cdot\text{m}^{-3}$), S&H ($3751.78 \pm 3732.87 \mu\text{g}\cdot\text{m}^{-3}$) and BJ ($511.89 \mu\text{g}\cdot\text{m}^{-3}$). The average MIR was ranging from 1.77 to 4.60 and did not show great difference between cuisine types.

Alkenes and aromatics exhibited high MIR values. The OFP from alkenes contributed most part of

OFP in our study. BBQ had both the highest OFP and alkene OFP (82%). The results were consistent with previous study that the sensitivity component of Home cooking, Shandong cuisine, and Barbecue were mainly contributed by alkenes, and the OFP accounted for 62.8%, 61.9%, and 75.6% of the total OFP, respectively (Cheng et al. 2016). Wang et al (2018) also revealed that the top contributor to OFP was alkenes for Canteen, Authentic Shanghai cuisine, Shaoxing cuisine and Cantonese cuisine. He et al (2020) proved that in terms of Family cuisine and Shandong cuisine, the sensitivity species were alkenes, and the OFP accounted for 51.7% and 56.6%. The main alkene of BBQ were 1-butene ($4205.33 \mu\text{g}\cdot\text{m}^{-3}$) and trans-2-butene ($11383.20 \mu\text{g}\cdot\text{m}^{-3}$). The reason of high butene emission maybe the incomplete combustion of carbon and fats (Umano et al. 1987). BBQ showed minimal aromatic OFP due to the low aromatic concentration of itself. The proportions of OFP from aromatics for S&H, HC, SD AH and BJ were similar (14%~29%). Notably, acrolein took roughly 30% in OVOCs for all cuisine types and exhibited high OFP proportion (53%~92%) of OVOCs OFP in our study.

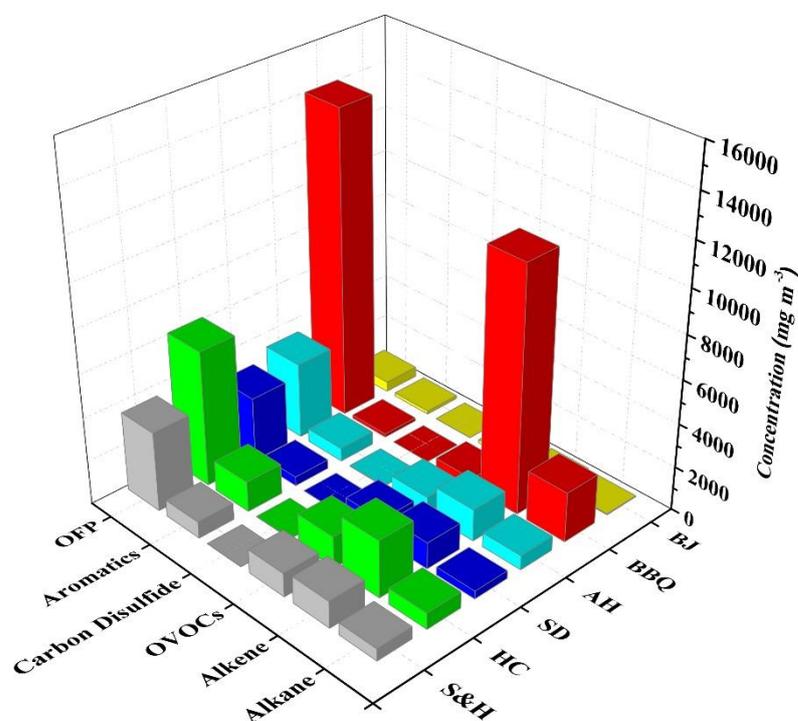


Fig.2. OFP characterizes of different cooking cuisines

Table.2. Ozone formation potential of VOCs emitted from different cooking cuisines

Components	S & H		HC		SD		AH*	BBQ*	BJ*
	avg	sd	avg	sd	avg	sd			
	$\mu\text{g}\cdot\text{m}^{-3}$		$\mu\text{g}\cdot\text{m}^{-3}$		$\mu\text{g}\cdot\text{m}^{-3}$		$\mu\text{g}\cdot\text{m}^{-3}$	$\mu\text{g}\cdot\text{m}^{-3}$	$\mu\text{g}\cdot\text{m}^{-3}$
Alkanes	519.78	526.14	755.91	1181.42	1229.18	429.99	1237.36	5012.45	78.45
Alkenes	1388.56	1462.97	2631.88	4891.91	1682.43	1242.02	1846.80	23800.42	114.02
OVOCS	1212.10	1336.85	1286.55	1314.77	1137.79	1046.99	1238.55	98.34	164.42
carbon disulfide	12.19	11.31	17.59	14.99	17.52	6.72	18.29	0.45	6.12
Aromatics	619.15	395.58	757.56	921.69	643.20	532.59	847.10	15.53	148.87
Total OFP	3751.78	3732.87	5449.48	8324.78	4710.10	3258.30	5188.10	28927.19	511.89
Average- MIR**	2.73	2.56	3.02	3.47	2.10	1.35	2.97	4.60	1.77

*The sample amount of this cuisine type was 1 and did not have a standard deviation. ** The units of average- MIR was $\text{g O}_3 \cdot \text{g VOCs}^{-1}$

Alkane contained: compound 1 ~ 11, 13, 15 ~ 20, 22 ~ 24 and 27 ~ 30; Alkene contained: compound 31 ~ 41. OVOCS contained: compound 44 ~ 48 and 50 ~ 55. Aromatics contained: compound 56 ~ 66 and 68 ~ 71.

4.3 Stench index of different cooking cuisine

In our research, the degree of stench pollution from cooking VOCs of HC was 17.51 ± 16.95 , followed by S&H (15.77 ± 16.85), SD (15.12 ± 14.17), AH (16.21), BBQ (2.57) and BJ (1.81) (Table 3). Except for OVOCS, the stench index of different cooking cuisines was similar to the conclusion of Shuiyuan et al. (2016) Notably, acrolein took the main part of the odor threshold and exist a certain degree of stench pollution. Considering the dilution effect of the ambient air toward the emitted gas, the VOCs from catering industry may had trivial contribution to the ambient stench pollution. However, the residents who lived nearby restaurants always complained about the stench pollution. We further calculated the stench pollution of ozone which came from OFP and the results showed a high degree of stench pollution. The stench pollution from the OFP of BBQ was 4218.55, following HC (1214.03 ± 686.89), AH (756.60), SD (686.89 ± 475.17), S&H (547.13 ± 544.38) and BJ (74.65). That maybe one of the reasons why the stench pollution from restaurants annoyed the residents.

Table 3 Stench index of VOCs components

Components	S&H		HC		SD		AH	BBQ	BJ
	avg	sd	avg	sd	avg	sd			
	$\mu\text{g}\cdot\text{m}^{-3}$		$\mu\text{g}\cdot\text{m}^{-3}$		$\mu\text{g}\cdot\text{m}^{-3}$		$\mu\text{g}\cdot\text{m}^{-3}$	$\mu\text{g}\cdot\text{m}^{-3}$	$\mu\text{g}\cdot\text{m}^{-3}$

Alkanes	0.14	0.09	0.13	0.13	0.16	0.08	0.23	0.48	0.02
Alkenes	0.30	0.29	0.39	0.51	0.31	0.15	0.34	0.74	0.04
OVOCs	14.15	15.92	16.20	15.77	13.63	13.30	14.96	1.35	1.29
Carbon disulfide	0.07	0.06	0.10	0.08	0.10	0.04	0.10	<0.01	0.03
Aromatics	1.12	0.49	0.68	0.45	0.81	0.38	0.66	<0.01	0.43
Halohydrocarbons	<0.01	<0.01	<0.01	<0.01	0.11	0.23	<0.01	<0.01	<0.01
TVOCs	15.77	16.85	17.51	16.95	15.12	14.17	16.29	2.58	1.81
Ozone	547.13	544.38	794.71	1214.03	686.89	475.17	756.60	4218.55	74.65

Alkene contained: compound 5, 7, 8, 11, 14, 17, 19, 22 ~ 24 and 27 ~ 30. Alkene contained: compound 32,33, 36, 38, 40 and 41. OVOCs contained: compound 44, 48, 51 and 52. Aromatics contained: compound 56 ~ 66, 69 and 70. Halohydrocarbons contained: compound 81 and 96.

4.4 Health risk assessment

According to *Exposure factors handbook of Chinese population (Adults)* (Xiuge and Xiaoli 2014), *ET*, *EF*, *ED*, *IR* and *AT* were assumed as $4 \text{ h}\cdot\text{d}^{-1}$, $365 \text{ d}\cdot\text{y}^{-1}$, 25 years, $0.65 \text{ m}^3\cdot\text{h}^{-1}$, 60 years (525600 h) for *LCR* and 25 years (219000h) for *HI*. The detailed data was described in **table 4**. The acceptable value of *LCR* and *HI* were 1.00×10^{-6} and 1(China 2019). The value of *RfC* and *IUR* were from USPEA-IRIS(USEPA 1992). In our research, the highest value of *LCR* occurred in the benzene, as for SD ($2.11\times 10^{-5} \pm 3.12\times 10^{-5}$) > HC ($4.50\times 10^{-6} \pm 3.83\times 10^{-6}$) > S&H ($4.08\times 10^{-6} \pm 4.49\times 10^{-6}$) > AH (4.13×10^{-6}). The *LCR* of tetrachloroethylene emission in SD was $7.57\times 10^{-6} \pm 1.50\times 10^{-5}$. The *LCR* of dichloromethane and vinyl chloride were acceptable.

Table 5 showed the *HI* for different cuisine types. Acrolein had great health risk and contributed 99% in *HI*. The *HI* of HC was 789.81 ± 768.77 , following S&H (689.89 ± 776.07), SD (664.29 ± 648.77), AH (728.78), BBQ (65.93) and BJ (62.84). The *HI* of tetrachloroethylene emission in SD was 1.747 ± 3.471 . The *HI* of benzene, toluene, dichloromethane, 2-hexanone and vinyl chloride were acceptable. Although acrolein was carcinogen, the *IUR* of acrolein had not been evaluated.

Table 4. *LCR* of different cuisine types

Components	<i>IUR</i> *	S & H		HC		SD		AH	BBQ	BJ
		avg	sd	avg	sd	avg	sd			

benzene	2.20×10^{-6}	4.08×10^{-6}	4.49×10^{-6}	4.50×10^{-6}	3.83×10^{-6}	2.11×10^{-5}	3.12×10^{-5}	4.13×10^{-6}	9.75×10^{-8}	4.98×10^{-7}
dichloromethane	1.00×10^{-8}	5.45×10^{-9}	4.81×10^{-9}	9.36×10^{-9}	7.77×10^{-9}	6.99×10^{-9}	1.66×10^{-9}	8.26×10^{-9}	8.91×10^{-10}	2.41×10^{-9}
tetrachloroethylene	2.60×10^{-7}	1.52×10^{-8}	1.34×10^{-8}	3.75×10^{-8}	6.86×10^{-8}	7.57×10^{-6}	1.50×10^{-5}	1.77×10^{-8}	9.43×10^{-10}	8.89×10^{-9}
vinyl chloride	4.40×10^{-6}	1.58×10^{-8}	1.54×10^{-8}	4.49×10^{-8}	6.18×10^{-8}	3.05×10^{-8}	2.14×10^{-8}	4.10×10^{-8}	0	4.36×10^{-9}
<i>Total LCR</i>		4.11×10^{-6}	4.53×10^{-6}	4.60×10^{-6}	3.97×10^{-6}	2.87×10^{-5}	4.62×10^{-5}	4.19×10^{-6}	9.93×10^{-8}	5.13×10^{-7}

*The unit of *IUR* was per $\text{mg} \cdot \text{m}^{-3}$

Table 5. *HI* of different cuisine types

Components	<i>RfC</i>	S & H		HC		SD		AH	BBQ	BJ
	$\text{mg} \cdot \text{m}^{-3}$	avg	sd	avg	sd	avg	sd			
cyclohexane	6.00	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01
acrolein	<0.01	689.89	776.07	789.81	768.73	664.29	648.77	728.78	65.93	62.84
benzene	0.03	0.15	0.16	0.16	0.14	0.77	1.13	0.15	<0.01	0.02
toluene	5.00	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01
dichloromethane	0.60	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01
trichloromethane	0.04	0.03	0.03	0.05	0.04	0.05	0.02	0.04	<0.01	0.01
tetrachloroethylene	0.04	<0.01	<0.01	<0.01	0.02	1.75	3.47	<0.01	<0.01	<0.01
2-hexanone	0.03	0.15	0.17	0.08	0.16	0.05	0.08	0.01	<0.01	0.03
vinyl chloride	0.10	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01
<i>HI</i>		690.23	776.44	790.12	769.11	666.91	653.47	729.00	65.93	63.00

5. Conclusion

6 cuisine types were selected as database. For TVOCs and OFP, the result showed that BBQ had the highest pollute ability in both TVOCs ($6287.61 \mu\text{g} \cdot \text{m}^{-3}$) and OFP ($28927.19 \mu\text{g} \cdot \text{m}^{-3}$). HC was in the second position. The TVOCs concentration and OFP of S & H, SD and AH were similar. Obviously, SD characterized by high halohydrocarbons proportion (33%). BJ had the lowest pollute ability in TVOCs ($288.81 \mu\text{g} \cdot \text{m}^{-3}$), OFP ($511.89 \mu\text{g} \cdot \text{m}^{-3}$). The standard derivations of TVOCs emission of each cuisine types were almost all greater than their average concentrations. Although each cuisine types had their own character flavors, these characters had less relative with their TVOCs emission. Alkene emission took 24% to 81% of OFP for all cuisine types. The degree of stench pollution from cooking VOCs of HC was 17.51 ± 16.95 , followed by AH (16.29), S&H (15.77 ± 16.85), SD (15.12 ± 14.17), BBQ (2.58) and

BJ (1.81). The secondary ozone came from OFP maybe the key pollutant that affect the ambient air quality. For health risk assessment, we found that benzene had high LCR and acrolein had high HI. Acrolein took the main part in both odor threshold and health risk assessment. Since acrolein, a typical aldosterone, had high environmental impact capacity, further study will concentrate on aldosterone.

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Consent to Publish: Not applicable.

Availability of data and materials: The datasets used and analyzed during the current study are available from the corresponding author on reasonable request.

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Declaration of competing interest: The authors declare that they have no competing interests

Author Contribution: Formal analysis and Writing were performed by Chengyi Sun. Investigation was performed by Liyun Zhao. Conceptualization and data Curation were performed by Dr. Huahua Bai and Guoao Li. Resources and Project administration were performed by Aijun Shi. Visualization was performed by Xue Chen. Funding acquisition was performed by Lei Nie. All authors read and approved the final manuscript.

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Figures

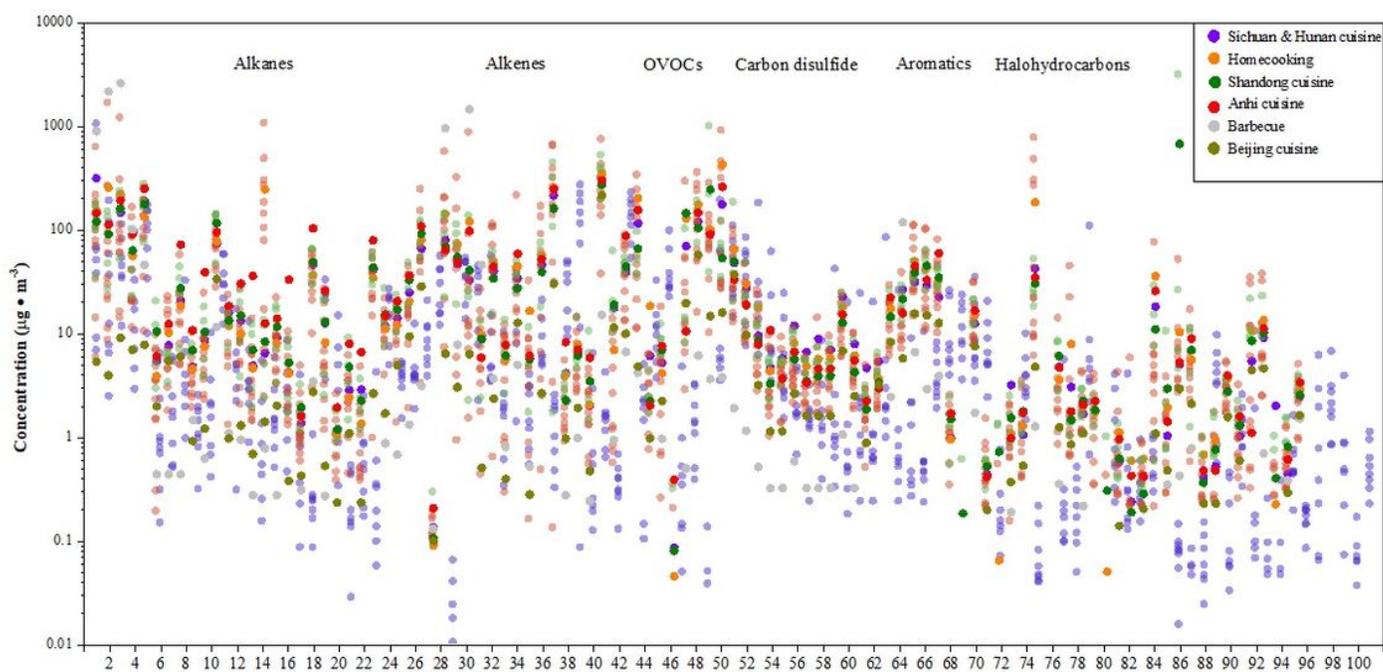


Figure 1

VOCs characterizes of different cooking cuisines (The abscissa axis was the serial numbers of compounds which were listed in table 1)

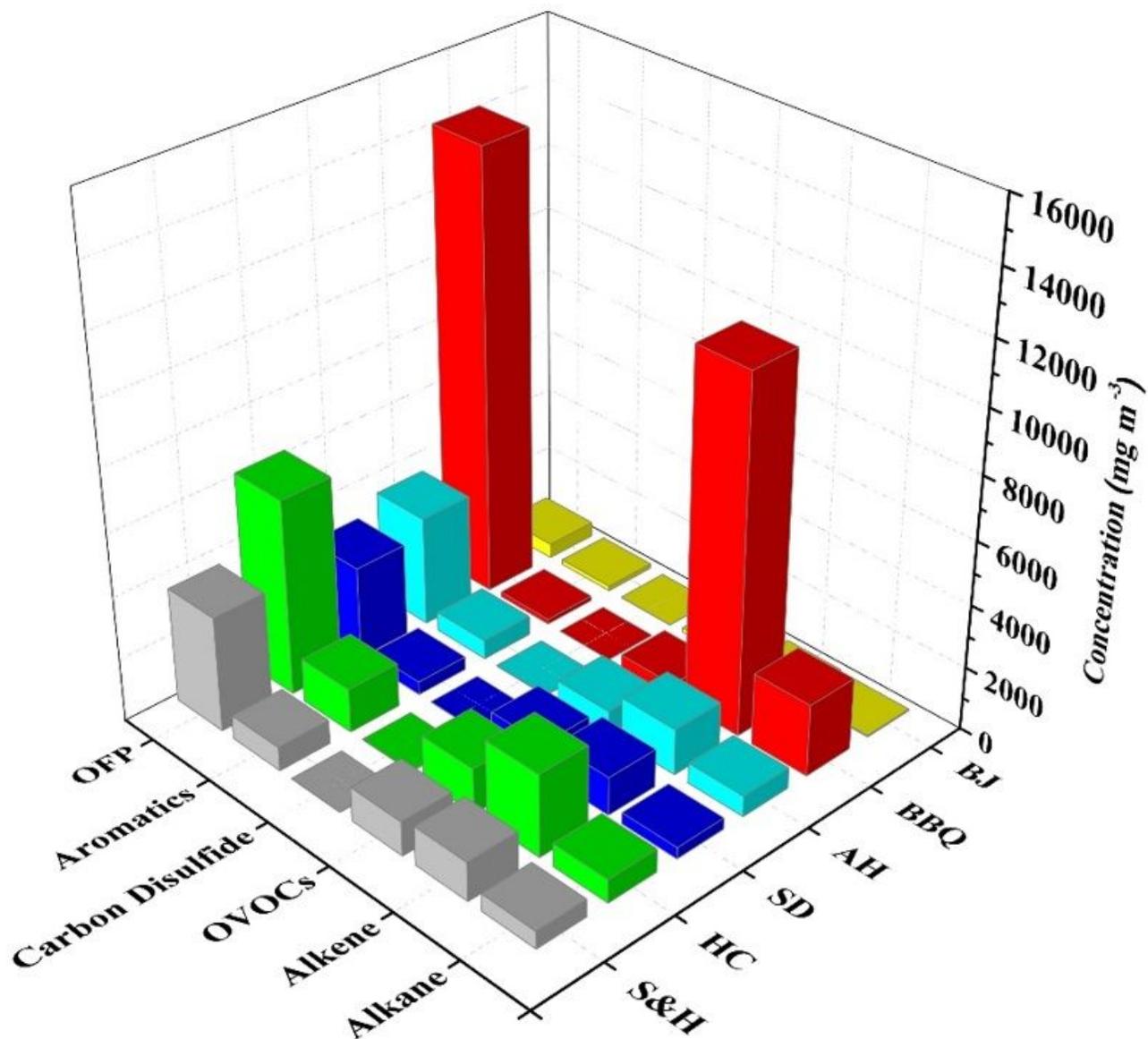


Figure 2

OFP characterizes of different cooking cuisines

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