

Available online at www.sciencedirect.com

ScienceDirect

www.elsevier.com/locate/jes

JES
 JOURNAL OF
 ENVIRONMENTAL
 SCIENCES
www.jesc.ac.cn

Chemical characterization of volatile organic compounds (VOCs) emitted from multiple cooking cuisines and purification efficiency assessments

Hanyu Zhang^{1,2}, Xuejun Wang^{1,2}, Xianbao Shen^{1,2}, Xin Li^{1,2}, Bobo Wu^{1,2}, Guohao Li³, Huahua Bai³, Xinyue Cao^{1,2}, Xuewei Hao^{1,2}, Qi Zhou^{1,2}, Zhiliang Yao^{1,2,*}

¹School of Ecology and Environment, Beijing Technology and Business University, Beijing 100048, China

²State Environmental Protection Key Laboratory of Food Chain Pollution Control, Beijing Technology and Business University, Beijing 100048, China

³Beijing Municipal Research Institute of Environmental Protection, Beijing Key Laboratory of Urban Atmospheric Volatile Organic Compounds Pollution Control and Application, National Urban Environmental Pollution Control Engineering Research Center, Beijing 100037, China

ARTICLE INFO

Article history:

Received 1 May 2022

Revised 25 July 2022

Accepted 2 August 2022

Available online 13 August 2022

Keywords:

Cooking emissions

Volatile organic compounds (VOCs)

VOCs categories

Ozone formation potential (OFP)

Purification efficiency

ABSTRACT

Cooking process can produce abundant volatile organic compounds (VOCs), which are harmful to environment and human health. Therefore, we conducted a comprehensive analysis in which VOCs emissions from multiple cuisines have been sampled based on the simulation and acquisition platform, involving concentration characteristics, ozone formation potential (OFP) and purification efficiency assessments. VOCs emissions varied from 1828.5 to 14,355.1 $\mu\text{g}/\text{m}^3$, with the maximum and minimum values from Barbecue and Family cuisine, respectively. Alkanes and alcohol had higher contributions to VOCs from Sichuan and Hunan cuisine (64.1%), Family cuisine (66.3%), Shandong cuisine (69.1%) and Cantonese cuisine (69.8%), with the dominant VOCs species of ethanol, isobutane and n-butane. In comparison, alcohols (79.5%) were abundant for Huaiyang cuisine, while alkanes (19.7%), alkenes (35.9%) and haloalkanes (22.9%) accounted for higher proportions from Barbecue. Specially, carbon tetrachloride, n-hexylene and 1-butene were the most abundant VOCs species for Barbecue, ranging from 8.8% to 14.6%. The highest OFP occurred in Barbecue. The sensitive species of OFP for Huaiyang cuisine were alcohols, while other cuisines were alkenes. Purification efficiency assessments shed light on the removal differences of individual and synergistic control technologies. VOCs emissions exhibited a strong dependence on the photocatalytic oxidation, with the removal efficiencies of 29.0%–54.4%. However, the high voltage electrostatic, wet purification and mechanical separation techniques played a mediocre or even counterproductive role in the VOCs reduction, meanwhile collaborative control technologies could not significantly improve the removal efficiency. Our results identified more effective control technologies, which were conducive to alleviating air pollution from cooking emissions.

© 2022 The Research Center for Eco-Environmental Sciences, Chinese Academy of Sciences. Published by Elsevier B.V.

* Corresponding author.

E-mail: yaozhl@th.btbu.edu.cn (Z. Yao).

Introduction

In recent years, people's requirements for diet have gradually increased as a consequence of the rapid development of economy and improvement of consumption levels. Diverse cuisines are flourishing, which are characterized by abundant cooking materials, unique seasoning, various cooking techniques based on multiple cuisines (Wang et al., 2018). However, a large amount of cooking lampblack rich in oil droplets, particulate matters (PMs), volatile organic compounds (VOCs) and other carcinogens (Huang et al., 2020; Zhang et al., 2017, 2021), were generated during the cooking process, which posed great threats to human health (Huang et al., 2011; Wang et al., 2017, 2018b). Especially for chefs, frequent exposure to heavy cooking lampblack, have a greater potential risk of lung cancer (Yu et al., 2015). Furthermore, VOCs from cooking emissions, as significant precursors of secondary organic aerosols and ozone, played a crucial role in the formation of aerosol pollution and photochemical smog (An et al., 2014; He et al., 2020). Numerous studies have demonstrated that cooking VOCs have become a critical contributor to ambient PMs and ozone, which deteriorated indoor and outdoor air quality considerably, and thus attracted more and more attention (Jung and Su, 2020; Ma et al., 2019; Sha et al., 2021). Therefore, it has become an urgent issue with respect to quantitative characterization of cooking VOCs and their components and effective removal of VOCs emissions by purification technology.

In the early stage, atmospheric environmental protection research in China mainly focused on the prevention and control of PMs, while the supervision of VOCs was relatively weak (Hao et al., 2016a, 2016b). In addition, considering the increasingly prominent O₃ pollution problem and the urgent need to meet refined environmental management in China (Li et al., 2020a; Wang et al., 2021), the VOCs from cooking emissions have been the focus of current attention and remediation (Li et al., 2020b; Pei et al., 2020). Correlational studies have revealed that more than 600 types of VOC chemical components could be generated during the cooking procedures, such as fatty acids, alkanes, alkenes, aromatics, alcohols, haloalkanes, aldehyde ketones, chlorides and heterocyclic compounds (Cheng et al., 2016; Wang et al., 2018; Huang et al., 2020; Kaltsonoudis et al., 2017; Klein et al., 2016; Liu et al., 2018). For instance, Cheng et al. (2016) only measured the four cuisine types of Home cooking, Shandong cuisine, Hunan cuisine and Barbecue, and detected alkanes, alkenes and aromatics, while the significant VOCs species were not considered, such as aldehydes and ketones, alcohol, and haloalkanes, and more cuisines need to be further compared. Peng et al. (2017) and Xiang et al. (2017) mainly focused on aldehydes and carbonyl compounds from cooking emissions, while more VOCs species were excluded, including alkanes, alkenes, aromatics, alcohol and haloalkanes. From what has been discussed above, the abovementioned studies, that have exclusively investigated the characteristics of a certain type or few types of VOC components from a limited number of cooking emissions, did not obtain and compare more comprehensive types of VOC components among multiple cuisines. Furthermore, systematic studies of the influence of cooking VOCs on the ozone formation potential are insuf-

ficient. He et al. (2020) and Kumar et al. (2018) demonstrated the ozone formation potential (OFP) of 32 VOC species (8 alkanes, 6 alkenes, 10 oxygenated-VOCs, 3 chlorinated VOCs and 5 benzene series) and 12 VOCs species (8 aromatics and 4 halogenated VOCs), respectively, and both identified the species that contributed most to OFP. However, the similarities and differences in the contributions of multiple sensitive species to OFP were not well described, and the number of species analyzed for OFP needed to be increased. Overall, exploring the whole species profiles of VOCs (covered alkanes, alkenes, aromatics, aldehydes, ketones, alcohol, haloalkanes, and etc.) generated from various cuisines and investigating the ozone formation potentials are thus urgent issues, which will allow for significant regulatory policy.

Moreover, how to effectively remove cooking lampblack has always been an urgent problem to be solved. Several purification technologies such as mechanical cleaning, electrostatic deposition, filtration adsorption, catalytic oxidation, photocatalytic oxidation, biodegradation and cold plasma technique have been developed and applied (Gysel et al., 2018a, 2018b; He et al., 2020; Holzer et al., 2018; Shu et al., 2018; Zielinska-Jurek and Zaleska, 2014). To our knowledge, most of the studies principally provided insights into the removal efficiency of the oil droplets and PMs in the cooking lampblack, whereas insights into removal efficiency of VOCs emitted from various cuisines were scarce and not systematic, particularly for various varieties of VOC species throughout the cooking process of multiple cuisines. Li et al. (2018) combined photocatalytic degradation and ozone oxidation purification techniques to remove cooking VOCs, and found that the maximum decomposition efficiencies of VOCs can reach up to 94%. Yao et al. (2019) achieved a 99.4% removal rate of hexanal from cooking lampblack using storage-plasma catalytic oxidation. Although previous researches have obtained substantial knowledge referring to the removal of change of cooking VOCs, there are terribly restricted comparative knowledge on the results of various removal technologies on the VOC species emitted from multiple cuisines.

Motivated by the aforementioned urgent needs, this study conducted lab measurements to reveal the concentration characteristics of VOCs and their species emitted from six cuisines, including alkanes, alkenes, aromatics, aldehydes and ketones, alcohols, haloalkanes, and others. The corresponding chemical reactivity of VOC compositions was also investigated. Furthermore, the similarities and variations of removal efficiency on the VOCs and their categories through multiple purification technologies were conjointly mentioned. The results were semiconducting to the promotion of environmental protection awareness and therefore the formulation of policies to mitigate pollution.

1. Materials and methods

1.1. Sampling methodology and collection

Based on the requirements of "Technical requirements and testing specifications for cooking fume purification equipment (trial)" (HJ/T62-2001), the cooking simulation and acquisition platform were established, which was designed to

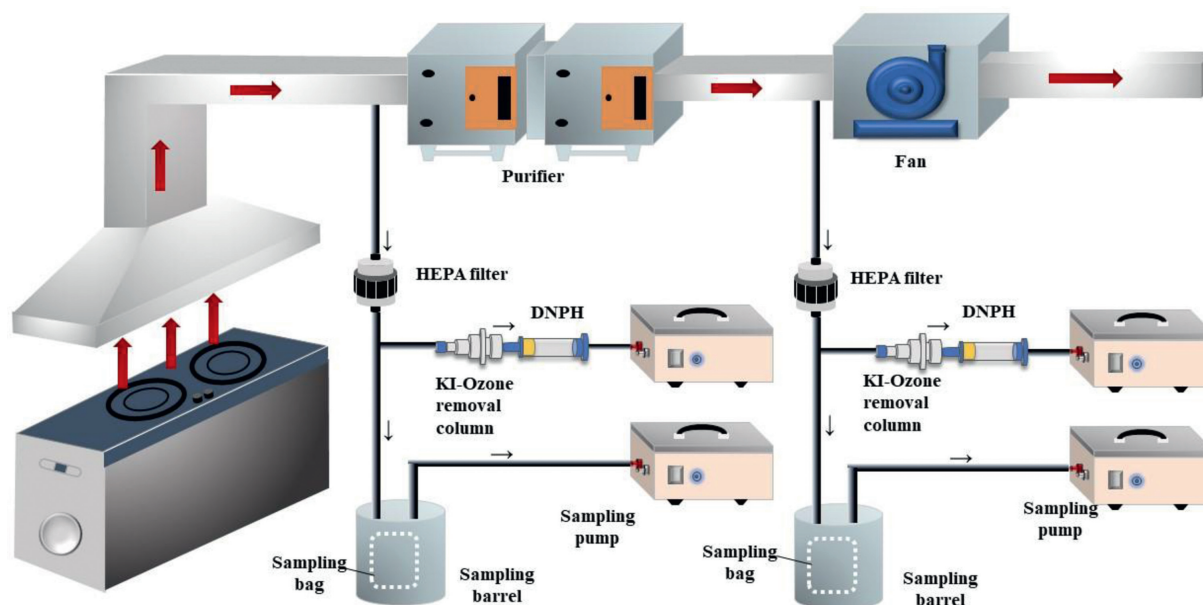


Fig. 1 – Schematic diagram of the cooking simulation and acquisition platform. HEPA: High Efficiency Particulate Air; DNPH: dinitrophenylhydrazine.

investigate pollutants emitted from cooking lampblack and evaluate the purification efficiency of different purification technologies. The schematic diagram of the platform setup is shown as Fig. 1. The sampling location was set within the straight section of the pipeline, which was set within the upstream direction not less than 1.5 times diameter and downstream direction not less than 3 times diameter from the elbow and the reducer. Six cuisine types were selected for sampling at the platform, including Shandong cuisine, Family cuisine, Sichuan and Hunan cuisine, Cantonese cuisine, Huaiyang cuisine and Barbecue, with the sampling time of 30 min. The detailed information on the cooking procedure is contained in Appendix A Table S1. A total of 92 VOCs were identified and quantified, of which 59 were analyzed in focus, consisting of 20 aldehydes and ketones, 10 alkanes, 10 alkenes, 8 aromatics, alcohol (ethanol), 5 haloalkanes, and 5 others. The detailed VOCs species were listed in Table 1.

Twenty aldehydes and ketones were collected by a silica cartridge impregnated with 2,4-dinitrophenylhydrazine (DNPH) (Sigma-Aldrich, Germany) at the flowrate of 1 L/min, when the cooking lampblack passed through the flue and purification system. The total cooking time for each cuisine were 30 min. In addition, Teflon filter and potassium iodide (KI) ozone removal columns were installed in front to remove particulate matter, and prevent the ozone in the pipeline from reacting with the derivative DNPH and the derived hydrazine, respectively. Soap film flowmeter were used to calibrate vacuum pump flow rate before sampling. All sampled cartridges were stored in laboratory refrigerators with Ultra Performance Liquid Chromatography (UPLC) before detection. Additionally, VOCs species other than aldehydes and ketones were sampled into tedlar bags by adopting stainless steel sampling gun under the negative pressure condition, and then were transported into the SUMMA canisters for storage and further de-

tection. The total flowrate was set to 0.5 L/min, and samples were collected for 30 min. To maximize the accuracy of sample concentration, SUMMA canisters, pipes as well as connections have been cleaned several times with ENTECH equipment and conducted vacuum backup before sampling. A more detailed description of the VOCs sampling can be found in the previous researches (Cheng et al., 2016; He et al., 2020; Wang et al., 2018).

1.2. Sample analysis

Each 2,4-DNPH silica cartridge for aldehydes and ketones analysis was extracted by means of 5.0 mL acetonitrile solution and injected into a 5.0 mL volumetric flask prior to the detection (Gysel et al., 2018b). The eluent was filtered with a 0.25 μm filter membrane and treated with an ultrasonic cleaner for 5 min. Acetonitrile was used for constant volume to the marking line. Collected aldehydes and ketones were thermally and analyzed by Ultra Performance Liquid Chromatographic instrument (Agilent 1290 Infinity LC, The United States) based on the UPLC method. The chromatographic column was a 2.1 \times 100 mm Hypersil ODS 1.8 μm reversed phase column (Zorbax RRHD Eclipse Plus C18, The United States) at room temperature. Certified calibration standards of TO11/IP-6A Aldehyde/Ketone-DNPH Mix were purchased from Supelco (Bellefonte, PA, The United States), with the concentration of 15,000 $\mu\text{g/L}$. The flow rate was 0.5 mL/min, and the column temperature was maintained at 40°C. The diode array detector (DAD) with a wavelength of 360 nm with the injection volume of 2 mL was employed to quantify the aldehydes and ketones. The gradient program is summarized in Table 2.

The other VOC species were transported into the automatic sampler by precooling enrichment system, which were quantitatively detected by Gas Chromatography-Mass Spectrometry.

Table 1 – VOC species detected in this study.

No.	Species	No.	Species	No.	Species
Aldehydes and ketones (20)					
1	Acetone	21	n-Butane	41	Benzene
2	2-Butanone	22	Isobutane	42	Toluene
3	Cyclohexanone	23	n-Pentane	43	Ethyl benzene
4	Formaldehyde	24	Isopentane	44	m/p-Xylene
5	Acetaldehyde	25	n-Hexane	45	o-Xylene
6	Acrolein	26	Cyclohexane	46	Styrene
7	Propionaldehyde	27	2-Methylpent	47	1,2,4-Trimethylbenzene
8	Crotonaldehyde	28	Heptane	48	p-Diethylbenzene
9	Butyl aldehyde	29	Octane	Alcohols (1)	
10	Benzaldehyde	30	2,4-Dimethylheptane	49	ethanol
Alkanes (10)					
11	Isovaleraldehyde	31	Propene	Haloalkanes (5)	
12	Valeraldehyde	32	1,3-Butadiene	50	Chloromethane
13	O-methyl benzaldehyde	33	1-Butene	51	Dichloromethane
14	m/p-Methyl benzaldehyde	34	cis-2-Butene	52	1,1-Dichloromethane
15	Hexanal	35	trans-2-Butene	53	Carbon tetrachloride
16	2,5-Dimethylbenzaldehyde	36	n-Pentene	54	1,2-Dichloroethane
17	Enanthaldehyde	37	trans-2-Pentene	Others (5)	
18	Caprylic aldehyde	38	Isoprene	55	Ethyl acetate
19	Nonanal	39	cis-2-Pentene	56	Tetrahydrofuran
20	Decanal	40	n-Hexylene	57	Chloroform
Aromatics (8)		Aromatics (8)		58	Propyl acetate
				59	Carbon disulfide

Table 2 – UPLC gradient elution program.

Elution Time/min	Ultrapure water (%)	Acetonitrile (%)	Mixture of water and tetrahydrofuran (%)
0	32	36	32
2.6	45	55	0
4	35	65	0
5.9	35	65	0
6.0	32	36	32
6.5	32	36	32

UPLC: Ultra Performance Liquid Chromatography.

try (Model 7890A/5975C, Agilent Inc, The United States) based on the U.S. EPA TO15 method (U.S. Environmental Protection Agency (EPA), 1999). Samples went through the first cryotrap (module 1) to eliminate water, N₂ and O₂ at –165°C by liquid nitrogen. The gaseous substances desorbed by module 1 were enriched and concentrated in a tenax adsorption trap at –50°C to remove Ar, CH₄, CO₂ and trace moisture through the second cryotrap (module 2) with the desorption temperature of 180°C. The samples would be frozen in the focused cold trap (module 3) with the freezing temperature of –160°C. And then module 3 rapidly heated up to vaporize the VOCs frozen in the capillary column head, driven by the carrier gas, desorbed into the capillary column of the chromatograph (GC) for separation and then entered the mass spectrometer (MS) for detection.

The chromatographic column was a DB-5MS (60 m × 0.35 mm × 1 μm), and the carrier gas was high purity helium at a flow velocity of 1.2 mL/min. The temperature of the injection port was 250°C, and the temperature of the column was maintained at 40°C for 8 min, and so was heated to 220°C at a rate of 30°C/min and lasted for 5 min. The MS was operated in the total ion scan mode, with the scan range and speed of

35–300 u and 1.44 scan/sec. Additional detailed information about analytical methods can be referred in the previous studies (Cheng et al., 2016; He et al., 2020; Lu et al., 2020).

1.3. Calculation of ozone formation potential

The OFP was calculated by taking into consideration the concentration and maximum increment reactivity (MIR) of every VOCs species (Carter, 2010; He et al., 2020; Venecek et al., 2018), which has been widely applied to assess the roles of VOCs species in ozone formation in recent years (Wei et al., 2022; Zhang et al., 2020). In this study, OFP method was performed to accurately reveal the cooking VOCs reactivities and evaluate the impact on outdoor ozone formation. Note that the detected VOCs concentrations were obtained directly from cooking emissions and did not take into account the physical and chemical reaction processes that diffuse into the atmosphere, so the calculated OFP may be overestimated. The detailed calculation formula is as follows (Cheng et al., 2016; Wang et al., 2018):

$$\text{OFP}_i = \text{VOCs}_i \times \text{MIR}_i \quad (1)$$

$$\text{OFF}_{ij} = f_{ij} \times \text{MIR}_i \quad (2)$$

where, OFF_i ($\mu\text{g}/\text{m}^3$) and VOC_i ($\mu\text{g}/\text{m}^3$) refer to OFF and the mass concentration of the i th VOCs species, respectively. MIR_i (gO_3/gVOC) represents the maximum incremental reactivity of i th VOCs species (Carter, 2010). Furthermore, OFF_{ij} (gO_3/gVOC) represents the mass of O_3 (g) generated from the i th species in 1 g VOCs for emission source j . And f_{ij} (%) represents the percentage of the i th species in the emission source j .

1.4. Purification equipment for removing VOCs from multiple cuisines

The purification equipment was placed between the inlet of the fume pipe and the fan, whose purification efficiency can be calculated through the distinction between the inlet and outlet VOCs concentrations of purification equipment. Four main techniques including high voltage electrostatic method (HVE), wet purification method (WPM), mechanical separation method (MSM) and photocatalytic oxidation (PCO) were adopted to remove VOCs from the cooking emissions. HVE is to produce an electrostatic field sufficient to ionize the gas under the action of high voltage direct current. The lampblack is charged through the electrostatic field and moves to the opposite polar plate, and is further adsorbed on the plate to form a layer of oil fume particles to purify oil fume. WPM mainly uses a washing liquid added with surfactants, lye, and emulsifiers to improve oil-water mixing to absorb oily particles and some gaseous pollutants, so as to achieve the purpose of transferring pollutants from the gas phase to the liquid phase. MSM is based on the sinking of oil fume particles under its own gravity to achieve a physical separation treatment of oil fume. PCO refers to the use of a special ultraviolet lamp with the wavelength of 185 nm to cut the oil molecule chain to form small oil molecules. At the same time, the ozone produced by the ultraviolet light and oxygen in the air is adopted to oxidize the small oil molecules to generate water, CO_2 and trace amount of white powder, so as to achieve the purpose of purifying oil fume. In this study, the application of the PCO consists of the installation of ultraviolet photolysis purification equipment in the gas hood and in the exhaust flue. Moreover, multiple combined purification technologies from different manufacturers were also applied and examined, as shown in Table 3. Based on the aforementioned methods, the purification efficiency of the total VOCs and VOCs species was investigated, respectively.

2. Results and discussion

2.1. Enhancements of VOCs from different cuisines

Overall, the total VOCs (TVOCs) concentrations for six cuisines decreased in the following order: Barbecue ($14,355.1 \pm 6894.6 \mu\text{g}/\text{m}^3$), Huaiyang cuisine ($3079.8 \pm 337.3 \mu\text{g}/\text{m}^3$), Sichuan and Hunan cuisine ($2680.0 \pm 495.5 \mu\text{g}/\text{m}^3$), Cantonese cuisine ($2283.7 \pm 706.7 \mu\text{g}/\text{m}^3$), Shandong cuisine ($2223.0 \pm 571.1 \mu\text{g}/\text{m}^3$), and Family cuisine ($1828.5 \pm 271.6 \mu\text{g}/\text{m}^3$), as illustrated in Fig. 2. It is noteworthy that the concentrations of TVOCs emitted from Barbecue were the

highest, 4.7–7.9 folds higher than the concentrations of TVOCs generated by other cuisines, which demonstrated open-air barbecues could exert a severe influence on air quality. This could attribute to inadequate combustion of charcoal, high temperature gasification of fat in meat and volatilization of sauce, which released more VOCs.

The investigated cuisines can be divided into four categories supported the VOC species. The VOCs emitted from Sichuan and Hunan cuisine showed nearly constant composition as Shandong cuisine, mainly composited by alkanes (31.1% and 36.1%) and alcohol (33.0% and 33.0%), respectively. Cooking wine, meat-derived fats and abundant edible oils may be also the explanation for the comparatively greater alkanes and alcohol production (He et al., 2020; Wang et al., 2018). Additionally, a larger amount concentration of alkanes was discovered, accounting for 48.1% and 46.1% with relevance Family cuisine and Cantonese cuisine, respectively. It is worth mentioning that alcohol from Huaiyang cuisine was super-high, which contributed about 80%. This is often in the main as a result of that rice wine or preparation wine were unre-markably adopted because the flavor once pickling or preparation some recent ingredients, and evaporative emissions of ethanol led to the high proportions of alcohol (Wang et al., 2018). Comparatively, alkenes and haloalkanes were the dominant VOCs contributors for Barbecue, with the proportion of 35.9% and 22.9%, respectively. The high alkenes concentration might be a consequence of enhanced edible oil burning and incomplete combustion of grease and charcoal. Chlorine disinfectants from water, detergents containing chloride, pesticide residues from the meat and meat chlorine washing process were possible reasons for the significant increase of aromatic haloalkanes (Masuda et al., 2020; Mattila et al., 2020).

2.2. Variations of VOCs profiles for each cuisine

Top ten VOC species were selected to demonstrate the pollution characteristics of key VOC species from six cuisines, as depicted in Fig. 3. For Sichuan and Hunan cuisine, ethanol comprised a large proportion of TVOCs, followed by the isobutane and n-butane, accounting for 33.0%, 12.7% and 12.3%, respectively. Note that the top ten VOCs produced by Shandong cuisine were similar to those of Sichuan and Hunan cuisine, especially for three significant species (containing ethanol, isobutane and n-butane), accounting for 33.0%, 15.8% and 14.5%, respectively. In terms of Family cuisine, the top ten VOC concentrations ranged from 19.5 to $427.5 \mu\text{g}/\text{m}^3$, accounting for 88.3% of the TVOCs. n-butane, isobutane and ethanol were the main contributors, with the proportion of 23.4%, 20.0% and 18.2%, respectively. With respect to Barbecue, the top ten VOCs species were comprised of carbon tetrachloride, n-hexylene, 1-butene, dichloromethane, n-pentene, propene, n-butane, benzene, n-pentane and heptane, with the concentrations ranging from 633.6 to $2097.9 \mu\text{g}/\text{m}^3$, accounting for 4.4%–14.6% of the TVOCs emissions. As far as Cantonese cuisine, ethanol, isobutane, n-butane and n-pentane were the main contributors, and made up the majority of the top ten VOCs species, accounting for 66.5% of TVOCs, with the percentage ranging from 9.1% to 23.7%. In contrast to the aforementioned cuisines, the top 10 VOCs species emitted by Huaiyang cuisine has significant difference, with the

Table 3 – Multiple combined purification technologies applied in various cuisines and corresponding VOC species detected.

Case	Purification technology	Cooking cuisine	VOC species detected
#1	HVE	Sichuan and Hunan cuisine	all species
#2		Sichuan and Hunan cuisine	all species
#3		Barbecue	aldehydes and ketones
#4	WPM	Huaiyang cuisine	all species
#5		Huaiyang cuisine	aldehydes and ketones
#6	MSM	Cantonese cuisine	aldehydes and ketones
#7		Sichuan and Hunan cuisine	all species
#8		Barbecue	aldehydes and ketones
#9	PCO-hood	Shandong cuisine	all species
#10		Shandong cuisine	all species
#11		Shandong cuisine	all species
#12		Sichuan and Hunan cuisine	all species
#13	PCO-flue	Sichuan and Hunan cuisine	all species
#14		Barbecue	aldehydes and ketones
#15	HVE+WPM	Family cuisine	all species
#16		Sichuan and Hunan cuisine	all species
#17		Sichuan and Hunan cuisine	all species
#18	HVE+MSM	Sichuan and Hunan cuisine	all species
#19		Barbecue	aldehydes and ketones
#20	HVE+PCO-flue	Sichuan and Hunan cuisine	all species
#21		Barbecue	aldehydes and ketones
#22	MSM+PCO-flue	Barbecue	aldehydes and ketones
#23	HVE+WPM+PCO-flue	Sichuan and Hunan cuisine	all species
#24		Barbecue	aldehydes and ketones

HVE: high voltage electrostatic method; WPM: wet purification method; MSM: mechanical separation method; PCO-hood: photocatalytic oxidation with hood; PCO-flue: photocatalytic oxidation with flue; HVE+WPM: high voltage electrostatic method coupled with wet purification method; HVE+MSM: high voltage electrostatic method coupled with mechanical separation method; HVE+PCO-flue: high voltage electrostatic method coupled with photocatalytic oxidation with flue; MSM+PCO-flue: mechanical separation method coupled with photocatalytic oxidation with flue; HVE+WPM+PCO-flue: high voltage electrostatic method coupled with wet purification method and photocatalytic oxidation with flue.

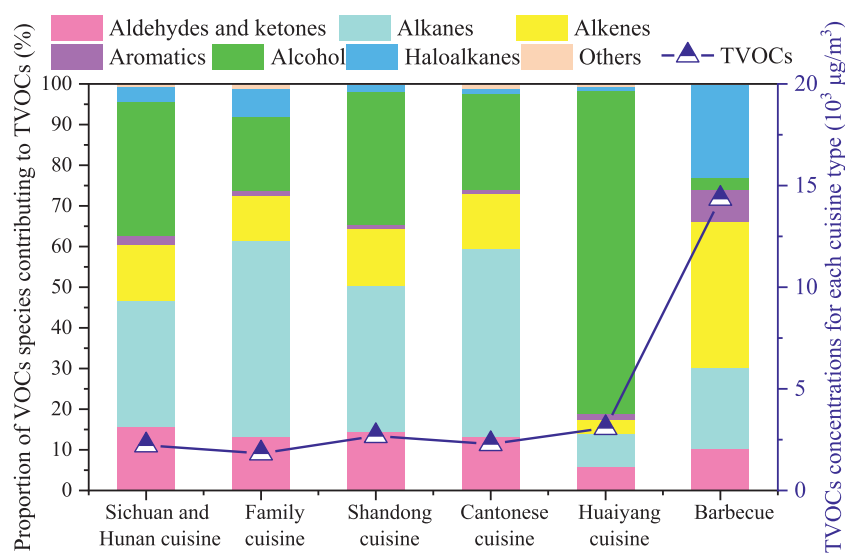


Fig. 2 – TVOC concentrations and proportion of VOC categories for each cuisine type. VOCs: volatile organic compounds; TVOCs: total volatile organic compounds.

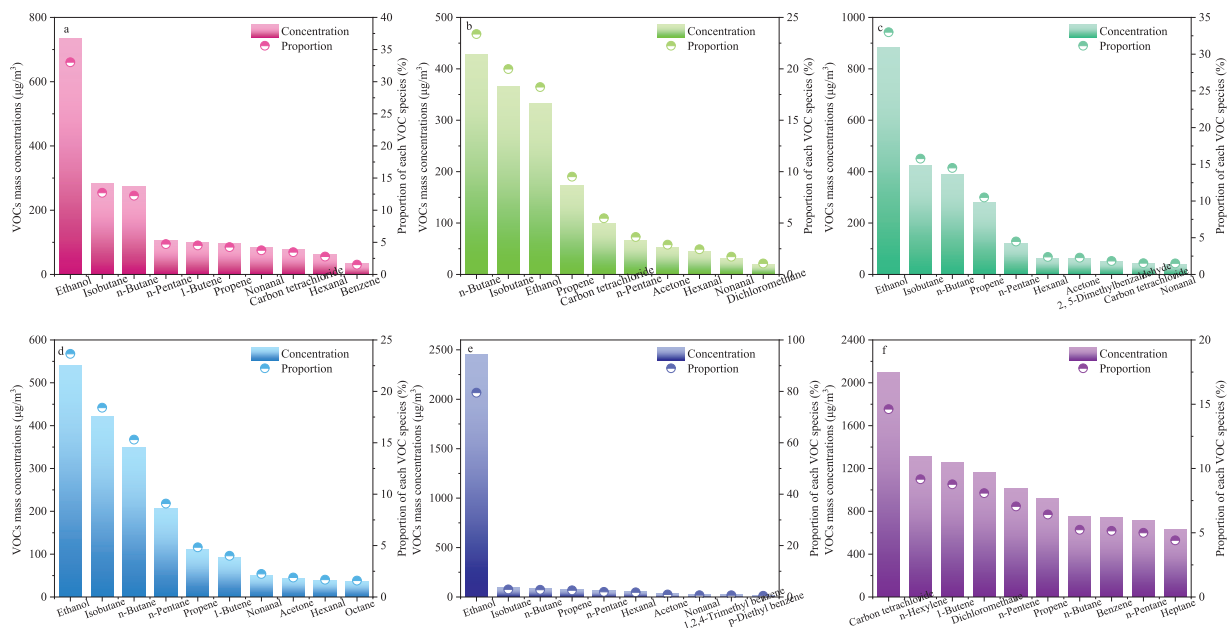


Fig. 3 – Top ten VOC species with the highest concentrations ($\mu\text{g}/\text{m}^3$) and corresponding percentages (%) from (a) Sichuan and Hunan cuisine, (b) Family cuisine, (c) Shandong cuisine, (4) Cantonese cuisine, (5) Huaiyang cuisine and (6) Barbecue.

ethylene concentration occupying an overwhelming proportion, contributing nearly 80%. Except for the differences of VOCs species from six cuisines, we also found some consistencies among different cuisine types. For the top ten VOCs species, n-butane, n-pentane and propene were all abundant in the analyzed cuisines, with the proportions of 2.8%–23.4%, 2.0%–9.1% and 2.6%–10.5%, respectively. Furthermore, in addition to Barbecue, ethanol, isobutane, 1-butene, nonanal and hexanal were the common dominant VOCs species of the other five cuisines. This suggested that consistencies and discrepancies coexist between the top ten VOCs profiles with the features of the different cuisines.

2.3. VOC reactivity evaluation

The ozone formation potential (OFP) of each category of VOCs for all cuisine types are shown in Fig. 4. The OFPs were ranked in order of Barbecue > Shandong cuisine > Cantonese cuisine > Sichuan and Hunan cuisine > Huaiyang cuisine > Family cuisine. It was noteworthy that the OFP of Barbecue was 6.9–11.8 times higher than those of the other five cuisines. Notably, the TVOCs concentrations of Huaiyang cuisine were 35% higher than that of Cantonese cuisine, while the OFP_i of two cuisines was comparable, manifesting that the cuisines with greater TVOCs concentrations may not necessarily produce greater potentials for ozone formation. Meanwhile, OFP_{ij} can be considered as the average OFP_i per unit mass of VOC, with the maximum value of $4.0 \text{ gO}_3/\text{gVOC}$ from Barbecue among all the cuisines. Compared with Huaiyang cuisine, although the TVOCs concentration and OFP_i of Family cuisine were relatively lower, the OFP_{ij} of Family cuisine was higher than that of Huaiyang cuisine due to the higher proportion and corresponding MIR of alkanes and alkenes in Family cuisine.

The contribution proportion of VOC species and categories from six cuisines to OFP have important variations, as presented in Fig. 5. With respect to Barbecue, the dominant VOC categories were alkenes, contributing 80.6% of the entire OFP. Though the characteristics of VOCs categories were different for Sichuan and Hunan cuisine, Family cuisine, Shandong cuisine and Cantonese cuisine, the contributions of the VOCs categories to their OFPs have similar distributions. In other words, alkenes were the most contributors to the OFP of aforementioned cuisines, accounting for 50.7%, 50.0%, 54.6% and 52.9%, respectively, followed by alkanes or alcohol. It was noteworthy that a high proportion of alcohol from Huaiyang cuisine were observed (60.6%), while the proportion of alkenes were solely 20.2%.

The top five VOC species to the OFP were selected to identify the key VOC species of OFP, with the total contribution rate ranging from 60.9% to 84.3%. For Sichuan and Hunan cuisine, the OFP contribution proportion of ethanol, propylene and butene was evenly distributed, reaching 17.8%, 17.4% and 15.4%, respectively. For Family cuisine, propene was the biggest contributor (41.8%) due to its higher MIR value and concentration, followed by ethanol (10.5%), n-butane (10.1%) and isobutane (9.3%). The OFP characteristics for Shandong cuisine were similar to those of Family cuisine; propene, ethanol, isobutane, cis-2-butene and n-butane were the top five primary VOCs species, contributing 39.6%, 16.3%, 6.3%, 5.7% and 5.4%, respectively. In the case of Cantonese cuisine, propene, 1-butene, ethanol and isobutane also showed higher percentages to the OFP, accounting for 20.2%, 14.0%, 13.0% and 8.1%, respectively. Owing to the higher concentration, ethanol became the dominant contributing specie to the OFP (60.6%), followed by propene (15.1%). The OFP characteristics of VOCs species

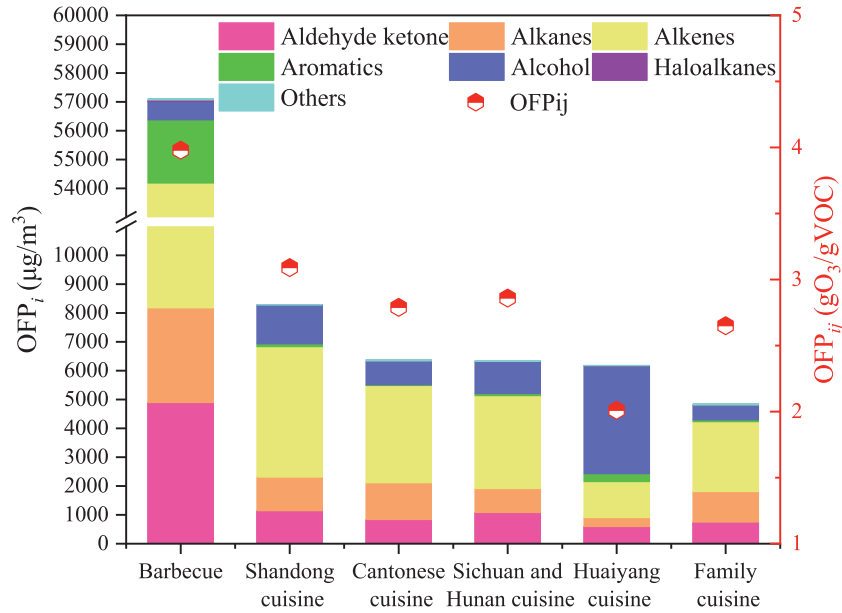


Fig. 4 – Contributions of different VOC categories produced by six cuisines to the ozone formation potential. OFP_i : ozone formation potential; OFP_{ij} : the mass of O_3 (g) generated from the i th species in 1 g VOCs for emission source j .

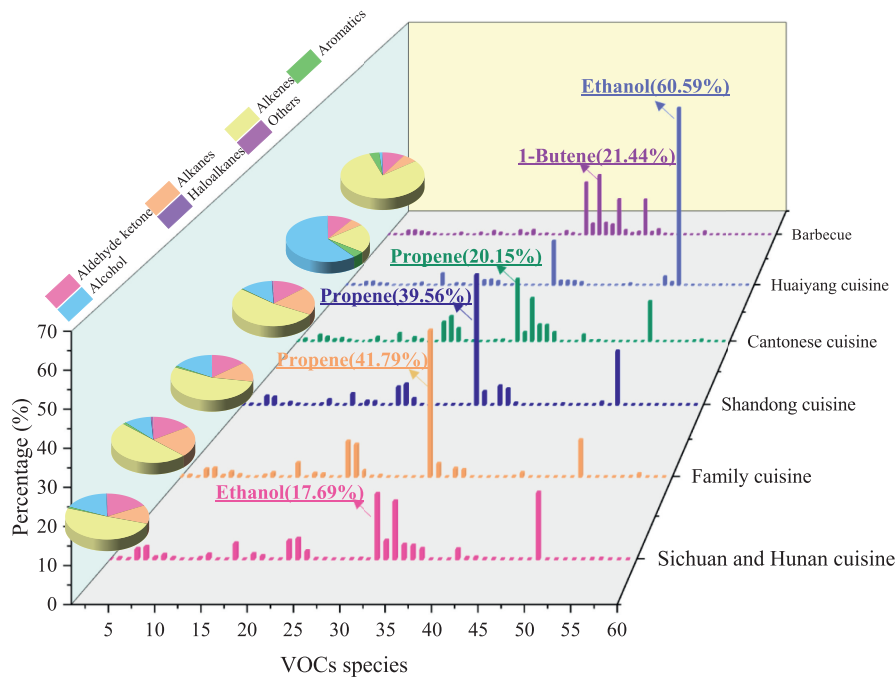


Fig. 5 – Contribution percentage of each VOC species and categories emitted from six cuisines to the ozone formation potential.

from Barbecue were notably different from other cuisines, showing the top five contributing VOCs species were all alkenes, with contribution rate of 1-butene, propene, n-pentene, and n-hexylene more than 12%. Therefore, to better understand the influence of VOCs emitted from cooking emissions on air quality, both VOCs concentrations and chemical activity should be taken into consideration comprehensive.

2.4. Purification efficiency assessments

In this study, the purification efficiencies of different control technologies for TVOC and VOC categories emitted from multiple cuisines were investigated and calculated based on cooking simulation and acquisition platform, including 24 dish samples, 5 individual control technologies and 5 synergistic control technologies tested in total (see detailed information

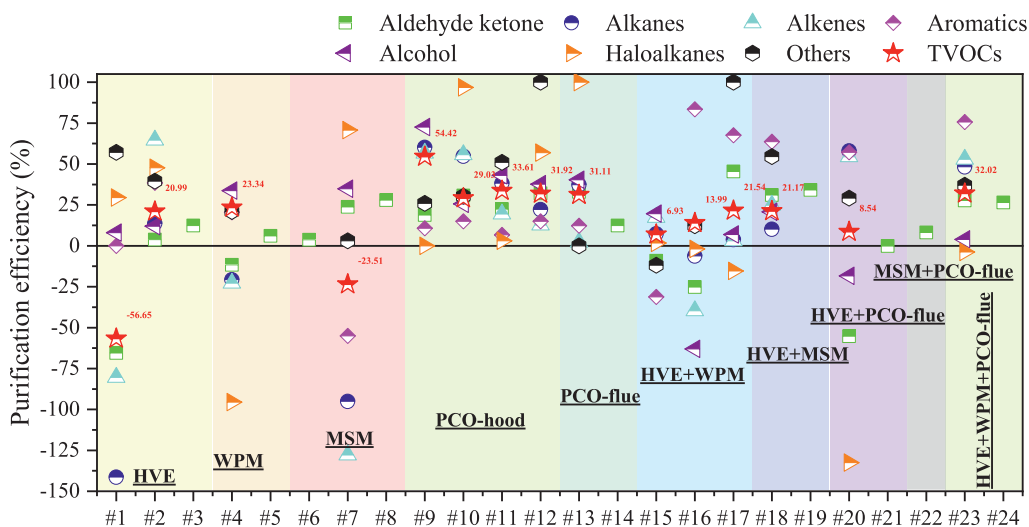


Fig. 6 – Purification efficiencies of different control technologies for TVOCs and VOCs categories emitted from multiple cuisines. HVE: high voltage electrostatic method; WPM: wet purification method; MSM: mechanical separation method; PCO-hood: photocatalytic oxidation with hood; PCO-flue: photocatalytic oxidation with flue; HVE+WPM: high voltage electrostatic method coupled with wet purification method; HVE+MSM: high voltage electrostatic method coupled with mechanical separation method; HVE+PCO-flue: high voltage electrostatic method coupled with photocatalytic oxidation with flue; MSM+PCO-flue: mechanical separation method coupled with photocatalytic oxidation with flue; HVE+WPM+PCO-flue: high voltage electrostatic method coupled with wet purification method and photocatalytic oxidation with flue.

in Table 3). As illustrated in Fig. 6, various control technologies presented a wide range of fluctuations for VOCs categories and TVOCs removal. HVE showed negative effects on the reduction of VOCs, with the removal efficiency of -56.7% for #1. It is worth noting that the increase of alkanes and alkenes in TVOCs emissions from #1 were 141.4% and 80.4% , respectively. This may be due to the fact that these newly formed micro molecular hydrocarbons were from the breaking of macro molecules. For WPM, the concentrations of TVOCs and alcohol in #4 have decreased dramatically, with the removal efficiencies of 23.3% and 33.7% , respectively. Normally, WPM mainly achieved the purpose of absorbing and removing VOCs by adding washing liquids (surfactants, lye and emulsifiers, etc.) with the function of improving oil-water mixing, which was mainly suitable for the removal of VOCs compounds with relatively single components. As a result, it thus effectively removed VOC from #4 (corresponding to Huaiyang cuisine), with the majority components of ethanol. In addition, the use of MSM exhibited increase in major VOCs categories (alkanes, alkenes and aromatics), with the TVOCs removal efficiencies of -23.5% . This could be attributed to the physical separation principle of MSM, which made it difficult to remove TVOCs. The HVE combined with WPM (HVE+WPM) and HVE combined with MSM (HVE+MSM) to remove VOCs in the oil fume were further explored. Overall, there is no significant improvement in purification efficiency. The removal efficiencies of TVOCs based on the aforementioned purification technologies were 6.9% (#15), 14.0% (#16), 21.5% (#17) and 21.2% (#18), respectively. The concentrations of some VOCs categories also increased, with the increase proportion of 25.2% (aldehydes

and ketones) and 39.7% (alkenes) from #16 by HVE+WPM, indicating instability in purification technology to a certain extent.

Notably, the adoption of photocatalytic oxidation control technology reflected obvious reductions in TVOCs and VOCs categories. In terms of PCO-hood and PCO-flue, the removal efficiencies of TVOCs were at ranges of $29.0\%–54.4\%$, much higher than those by HVE, WPM and MSM. Meanwhile, large emissions reductions were also found for major VOCs categories, with aldehydes and ketones, alkanes, alkenes and alcohol showing reduction proportions ranged from $12.4\%–32.9\%$, $22.1\%–60.2\%$, $2.3\%–56.7\%$ and $25.7\%–72.5\%$, respectively. The purification mechanism of the PCO-hood and PCO-flue is that most high-molecular weight VOCs could be transformed to low-molecular weight VOCs and further oxidized by ozone to CO_2 and H_2O under the action of ultraviolet light. Therefore, much more numerous VOCs categories could be oxidized and removed during the photocatalytic oxidation process. What is more, the synergistic removal efficiencies of HVE+PCO-flue and MSM+PCO-flue were lower than that of PCO-flue alone, with the reduction proportion of 8.5% for TVOCs and 8.1% for aldehydes and ketones. It is worth to be mentioned that the synergistic control technology of HVE+WPM+PCO-flue still remained higher removal efficiencies, with the reduction proportion of TVOCs and major VOCs categories higher than 25% . From what has been discussed above, PCO-hood and PCO-flue appeared to be the most effective technology followed by HVE+WPM+PCO-flue, with HVE and MSM being the least effective technologies in reducing TVOCs.

3. Conclusions

We investigated cooking VOC emissions from six types of cuisines in China including the characteristics of VOCs and their species, VOC reactivity evaluation moreover as purification efficiencies of multiple control technologies. The primary conclusions are summarized below:

Our results showed that TVOC mass concentrations were in the order of Barbecue ($14,355.1 \pm 6894.6 \mu\text{g}/\text{m}^3$) > Huaiyang cuisine ($3079.8 \pm 337.3 \mu\text{g}/\text{m}^3$) > Sichuan and Hunan cuisine ($2680.0 \pm 495.5 \mu\text{g}/\text{m}^3$) > Cantonese cuisine ($2283.7 \pm 706.7 \mu\text{g}/\text{m}^3$) > Shandong cuisine ($2223.0 \pm 571.1 \mu\text{g}/\text{m}^3$) > Family cuisine ($1828.5 \pm 271.6 \mu\text{g}/\text{m}^3$). Alkanes and alcohol were the main VOCs categories with respect to the Sichuan and Hunan cuisine, Family cuisine, Shandong cuisine and Cantonese cuisine, accounting for 31.1%–48.1% and 18.2%–33.0%, respectively. What is more, ethanol, isobutane and n-butane had the relatively higher contributions to these four cuisine types, with the proportion ranging from 18.2%–33.0%, 12.7%–20.0% and 12.3%–23.4%, respectively. Comparatively speaking, alcohol (ethanol) was the absolute dominant contributor to the Huaiyang cuisine (79.5%), whereas the abundance of alkanes, alkenes and haloalkanes produced from Barbecue were relatively higher, with the proportion of 19.7%, 35.9% and 22.9%, respectively. Specially, carbon tetrachloride, n-hexylene and 1-butene were the most abundant VOCs species for Barbecue, accounting for 14.6%, 9.2% and 8.8% of the TVOCs, respectively. In addition, TVOCs from Barbecue has the largest OFP, with the higher contributions from 1-butene (21.4%), propene (18.8%), n-pentene (12.8%) and n-hexylene (12.7%). In comparison, there were not too big differences of OFPs among the other cuisine types, falling in the range of 4850.3–8285.9 $\mu\text{g}/\text{m}^3$. The sensitivity compounds of OFP were propene and ethanol, accounting for 15.1%–41.8% and 10.5%–60.6%, respectively. Therefore, open-air barbecue should be taken seriously for its potential ozone pollution hazard, and the influence of other cuisines on ozone formation should not be ignored.

To better understand the effects of various control technologies on cooking VOC reductions, ten types of individual and synergistic control technologies were considered to assess the purification efficiencies of VOCs and VOCs categories emitted from six cuisine types. For HVE and MSM, instead of being effectively removed, the concentrations of most VOC categories have increased to a certain extent, suggesting the removal effects of these two control technologies were very limited. In comparison, the purification effects of WPM were slightly improved, resulting in a 23.3% reduction of the TVOC emissions. Furthermore, the removal effect of collaborative purification technology was not significantly improved, with removal efficiencies of TVOCs of 6.9% and 14.0% for HVE+WPM, and 21.5% and 21.2% for HVE+MSM, respectively. It is noteworthy that PCO-hood and PCO-flue has been proved to be significantly more efficient in removing cooking VOCs, with the removal efficiencies of TVOCs of 29.0%–54.4%, much higher than those by HVE, WPM, MSM and collaborative technologies among them. Nevertheless, the removal effects of TVOCs were not improved dramatically through the

combinations of PCO-flue with HVE, WPM or MSM. As a result, PCO-hood and PCO-flue should be promoted for cooking VOCs removal.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Acknowledgments

This work was supported by the Open Research Fund Program of State Environmental Protection Key Laboratory of Food Chain Pollution Control (No. FC2021YB03), the Research Foundation for Youth Scholars of Beijing Technology and Business University (No. QNJ2021-32). The authors are grateful to the anonymous reviewers for their insightful comments.

Appendix A Supplementary data

Supplementary material associated with this article can be found, in the online version, at doi:10.1016/j.jes.2022.08.008.

REFERENCES

- An, T.C., Huang, Y., Li, G.Y., He, Z.G., Chen, J.Y., Zhang, C.S., 2014. Pollution profiles and health risk assessment of VOCs emitted during e-waste dismantling processes associated with different dismantling methods. *Environ. Int.* 73, 186–194.
- Carter, W.P.L., 2010. Development of a condensed SAPRC-07 chemical mechanism. *Atmos. Environ.* 44, 5336–5345.
- Cheng, S.Y., Wang, G., Lang, J.L., Wen, W., Wang, X.Q., Yao, S., 2016. Characterization of volatile organic compounds from different cooking emissions. *Atmos. Environ.* 145, 299–307.
- Gysel, N., Dixit, P., Schmitz, D.A., Engling, G., Cho, A.K., Cocker, D.R., et al., 2018a. Chemical speciation, including polycyclic aromatic hydrocarbons (PAHs), and toxicity of particles emitted from meat cooking operations. *Sci. Total Environ.* 633, 1429–1436.
- Gysel, N., Welch, W.A., Chen, C.L., Dixit, P., Cocker, D.R., Karavalakis, G., 2018b. Particulate matter emissions and gaseous air toxic pollutants from commercial meat cooking operations. *J. Environ. Sci.* 65, 162–170.
- Hao, X.W., Li, J., Yao, Z.L., 2016a. Changes in PAHs levels in edible oils during deep-frying process. *Food Control* 66, 233–240.
- Hao, X.W., Yin, Y., Feng, S.J., Du, X., Yu, J.Y., Yao, Z.L., 2016b. Characteristics of polycyclic aromatic hydrocarbons in food oils in Beijing catering services. *Environ. Sci. Pollut. Res.* 23, 24932–24942.
- He, W.Q., Shi, A.J., Shao, X., Nie, L., Wang, T.Y., Li, G.H., 2020. Insights into the comprehensive characteristics of volatile organic compounds from multiple cooking emissions and aftertreatment control technologies application. *Atmos. Environ.* 240, 117646.
- Holzer, F., Kopinke, F.D., Roland, U., 2018. Non-thermal plasma treatment for the elimination of odorous compounds from exhaust air from cooking processes. *Chem. Eng. J.* 334, 1988–1995.

- Huang, X.Q., Han, D.M., Cheng, J.P., Chen, X.J., Zhou, Y., Liao, H.X., et al., 2020. Characteristics and health risk assessment of volatile organic compounds (VOCs) in restaurants in Shanghai. *Environ. Sci. Pollut. Res.* 27, 490–499.
- Huang, Y., Ho, S.S.H., Ho, K.F., Lee, S.C., Yu, J.Z., Louie, P.K.K., 2011. Characteristics and health impacts of VOCs and carbonyls associated with residential cooking activities in Hong Kong. *J. Hazard. Mater.* 186, 344–351.
- Jung, C.C., Su, H.J., 2020. Chemical and stable isotopic characteristics of $PM_{2.5}$ emitted from Chinese cooking. *Environ. Pollut.* 267, 115577.
- Kaltsonoudis, C., Kostenidou, E., Louvaris, E., Psichoudaki, M., Tsigliannis, E., Florou, K., et al., 2017. Characterization of fresh and aged organic aerosol emissions from meat charbroiling. *Atmos. Chem. Phys.* 17, 7143–7155.
- Klein, F., Platt, S.M., Farren, N.J., Detournay, A., Bruns, E.A., Bozzetti, C., et al., 2016. Characterization of gas-phase organics using proton transfer reaction time-of-flight mass spectrometry: cooking emissions. *Environ. Sci. Technol.* 50, 1243–1250.
- Kumar, A., Singh, D., Kumar, K., Singh, B.B., Jain, V.K., 2018. Distribution of VOCs in urban and rural atmospheres of subtropical India: temporal variation, source attribution, ratios, OFP and risk assessment. *Sci. Total Environ.* 613, 492–501.
- Li, J.X., Wang, Z.X., Chen, L.L., Lian, L.L., Li, Y., Zhao, L.Y., et al., 2020a. WRF-Chem simulations of ozone pollution and control strategy in petrochemical industrialized and heavily polluted Lanzhou City, Northwestern China. *Sci. Total Environ.* 737, 115577.
- Li, Y.H., Cheng, S.W., Yuan, C.S., Lai, T.F., Hung, C.H., 2018. Removing volatile organic compounds in cooking fume by nano-sized TiO_2 photocatalytic reaction combined with ozone oxidation technique. *Chemosphere* 208, 808–817.
- Li, Z.Y., Ho, K.F., Yim, S.H.L., 2020b. Source apportionment of hourly-resolved ambient volatile organic compounds: in fluence of temporal resolution. *Sci. Total Environ.* 725, 138243.
- Liu, T.Y., Wang, Z.Y., Huang, D.D., Wang, X.M., Chan, C.K., 2018. Significant production of secondary organic aerosol from emissions of heated cooking oils. *Environ. Sci. Technol. Lett.* 5, 32–37.
- Lu, F.J., Li, S.H., Shen, B.X., Zhang, J.W., Liu, L.J., Shen, X.Q., et al., 2020. The emission characteristic of VOCs and the toxicity of BTEX from different mosquito-repellent incenses. *J. Hazard. Mater.* 384, 121428.
- Ma, Z.B., Liu, C.T., Zhang, C.L., Liu, P.F., Ye, C., Xue, C.Y., 2019. The levels, sources and reactivity of volatile organic compounds in a typical urban area of Northeast China. *J. Environ. Sci.* 79, 121–134.
- Masuda, M., Wang, Q., Tokumura, M., Miyake, Y., Amagai, T., 2020. Risk assessment of polycyclic aromatic hydrocarbons and their chlorinated derivatives produced during cooking and released in exhaust gas. *Ecotoxicol. Environ. Saf.* 197, 110592.
- Mattila, J.M., Arata, C., Wang, C., Katz, E.F., Abeleira, A., Zhou, Y., et al., 2020. Dark chemistry during bleach cleaning enhances oxidation of organics and secondary organic aerosol production indoors. *Environ. Sci. Technol. Lett.* 7, 795–801.
- Pei, J.J., Yin, Y.H., Liu, J.J., Dai, X.L., 2020. An eight-city study of volatile organic compounds in Chinese residences: compounds, concentrations, and characteristics. *Sci. Total Environ.* 698, 134137.
- Peng, C.Y., Lan, C.H., Lin, P.C., Kuo, Y.C., 2017. Effects of cooking method, cooking oil, and food type on aldehyde emissions in cooking oil fumes. *J. Hazard. Mater.* 324, 160–167.
- Sha, Q.E., Zhu, M.N., Huang, H.W., Wang, Y.Z., Huang, Z.J., Zhang, X.C., et al., 2021. A newly integrated dataset of volatile organic compounds (VOCs) source profiles and implications for the future development of VOCs profiles in China. *Sci. Total Environ.* 793, 148348.
- Shu, Y.J., Ji, J., Xu, Y., Deng, J.G., Huang, H.B., He, M., et al., 2018. Promotional role of Mn doping on catalytic oxidation of VOCs over mesoporous TiO_2 under vacuum ultraviolet (VUV) irradiation. *Appl. Catal., B* 220, 78–87.
- U.S. Environmental Protection Agency (EPA), 1999. Compendium method TO-15 determination of volatile organic compounds (VOCs) in air collected in specially-prepared canisters and analyzed by GC/MS.
- Venecek, M.A., Carter, W.P.L., Kleeman, M.J., 2018. Updating the SAPRC maximum incremental reactivity (MIR) scale for the United States from 1988 to 2010. *J. Air Waste Manag. Assoc.* 68, 1301–1316.
- Wang, H.L., Xiang, Z.Y., Wang, L.N., Jing, S.A., Lou, S.R., Tao, S.K., et al., 2018. Emissions of volatile organic compounds (VOCs) from cooking and their speciation: a case study for Shanghai with implications for China. *Sci. Total Environ.* 621, 1300–1309.
- Wang, L.N., Xiang, Z.Y., Stevanovic, S., Ristovski, Z., Salimi, F., Gao, J., et al., 2017. Role of Chinese cooking emissions on ambient air quality and human health. *Sci. Total Environ.* 589, 173–181.
- Wang, L.N., Zheng, X.R., Stevanovic, S., Wu, X., Xiang, Z.Y., Yu, M.Z., et al., 2018b. Characterization particulate matter from several Chinese cooking dishes and implications in health effects. *J. Environ. Sci.* 72, 98–106.
- Wang, X.L., Fu, T.M., Zhang, L., Cao, H.S., Zhang, Q., Ma, H.C., et al., 2021. Sensitivities of ozone air pollution in the Beijing-Tianjin-Hebei area to local and upwind precursor emissions using adjoint modeling. *Environ. Sci. Technol.* 55, 5752–5762.
- Wei, W., Wang, X.F., Wang, X.Q., Li, R., Zhou, C.Y., Cheng, S.Y., 2022. Attenuated sensitivity of ozone to precursors in Beijing-Tianjin-Hebei region with the continuous NO_x reduction within 2014–2018. *Sci. Total Environ.* 813, 152589.
- Xiang, Z.Y., Wang, H.L., Stevanovic, S., Jing, S.A., Lou, S.R., Tao, S.K., et al., 2017. Assessing impacts of factors on carbonyl compounds emissions produced from several typical Chinese cooking. *Build. Environ.* 125, 348–355.
- Yao, X., Gao, M.X., Wei, Z.D., Chen, M.X., Shangguan, W.F., 2019. Removal of hexanal in cooking fume by combination of storage and plasma-catalytic oxidation on alkali-modified Co-Mn solid solution. *Chemosphere* 220, 738–747.
- Yu, K.P., Yang, K.R., Chen, Y.C., Gong, J.Y., Chen, Y.P., Shih, H.C., et al., 2015. Indoor air pollution from gas cooking in five Taiwanese families. *Build. Environ.* 93, 258–266.
- Zhang, N., Han, B., He, F., Xu, J., Zhao, R.J., Zhang, Y.J., et al., 2017. Chemical characteristic of $PM_{2.5}$ emission and inhalational carcinogenic risk of domestic Chinese cooking. *Environ. Pollut.* 227, 24–30.
- Zhang, X.M., Wang, D., Liu, Y., Cui, Y.F., Xue, Z.G., Gao, Z.F., et al., 2020. Characteristics and ozone formation potential of volatile organic compounds in emissions from a typical Chinese coking plant. *J. Environ. Sci.* 95, 183–189.
- Zhang, Z.R., Zhu, W.F., Hu, M., Wang, H., Chen, Z., Shen, R.Z., et al., 2021. Secondary organic aerosol from typical Chinese domestic cooking emissions. *Environ. Sci. Technol. Lett.* 8, 24–31.
- Zielinska-Jurek, A., Zaleska, A., 2014. Ag/Pt-modified TiO_2 nanoparticles for toluene photooxidation in the gas phase. *Catal. Today* 230, 104–111.