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# Assessing impacts of factors on carbonyl compounds emissions produced from several typical Chinese cooking



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# ABSTRACT

Carbonyl compounds are important constituents of photochemical smog and important precursors of photochemical smog formation. In order to investigate concentrations and types of carbonyls generated during cooking process, the influence of cooking styles, food materials, oil types and purification facilities were investigated. Silica cartridge impregnated with 2,4-dinitrophenylhydrazine (2,4-DNPH) and high performance liquid chromatography (HPLC) were adopted for collecting and analyzing samples. Results showed that the emission factors (EFs) of carbonyls (C1-C8) produced from cooking emissions varied within the range of 0.669-1.596 µg/kg. In relation to cooking styles, Barbecue (1.596 µg/kg) > Frying (1.530 µg/kg) > Teppanyaki (1.229 µg/kg) > Stir-frying (0.699 µg/kg), while C1-C3 carbonyls accounted for over 85% for the carbonyls concentrations. Regarding the food materials, generally, carbonyl emissions from meat dishes were greater than for non-meat dishes. Among different oil types used, cooking with sunflower oil resulted in the highest carbonyl emissions. In addition, carbonyls produced from frying carried the highest health risk. The high-voltage electrostatic methodology applied to remove carbonyls did not show as efficient for this purpose. Formaldehyde was found to be the largest contributor to ozone formation potential (OFP).

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## 1. Introduction

Carbonyl compounds are a group of chemically active volatile organic compounds (VOCs) involved in many atmospheric, photochemical reactions, such as ozone and peroxyacetyl nitrate (PANs) formation [1]. Carbonyls are of critical importance as products of photo-oxidation from gas-phase hydrocarbons, which are a major source of free radicals and the precursors to organic-aerosol formation in urban air. In addition, carbonyls have received increasing attention for their adverse health effects of irritants and carcinogens [2–5].

Cooking fume is one of the major sources of carbonyl compounds [6]. On one hand, cooking fuels, including charcoal and wood, could produce carbonyls during combustion [3,7]. On the other hand, pyrolysis and hydrolysis of edible oils and food materials at high temperatures can also produce carbonyls [8]. However, GB/T18883-2002 of China standard for indoor air only regulates the emitted concentrations of formaldehyde (0.10 mg/m<sup>3</sup>.h), and not for other compounds present.

There are two main analytical methodologies used for sampling and subsequent analysis of carbonyls: One includes collection on dinitrophenylhydrazine (DNPH) silica cartridges, followed by a high performance liquid chromatography (HPLC) used for analysis. The other one is using pentafluorobenzyl hydroxylamine (PFBHA) as a derivative of the adsorption column for sampling, and then gas chromatography-mass spectrometer (GC-MS) [9,10]. Current studies were mainly conducted in real restaurants [2,11–13]. It was found that fast food restaurants contribute the most to the total carbonyl emissions, whereas deep-fry cooking was the main contributor. Cooking with rapeseed salad oil produced the largest



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amount of carbonyls in comparison to other oil types. Few studies evaluated factors influencing levels of carbonyl emission in the laboratory, where the amount of food and oil, as well as ventilation conditions were controlled during cooking process.

This study conducted lab measurements to investigate carbonyl concentrations emitted from several typical Chinese cooking dishes using the most common food material and oil types being impacted by factors including cooking styles, oil types, food materials, and purification facilities. In addition, we investigated the role carbonyls have on the environment. Its ozone formation potential (OFP) was investigated, which was further used to assess the impact of VOCs on ozone formation for indoor and outdoor air [14]. Health impact of exposure to carbonyls produced during cooking was also assessed.

#### 2. Methodology

#### 2.1. Sample collections and analysis

The amount of food material (5 kg), oil consumption (200 ml), oil temperature (260 °C) and cooking time (1 h) were controlled strictly. Surface water content was maintained constant for each test by spraying the same amount of water after drying of food materials. The measurement setup is shown as Fig. 1.

When the cooking starts, cooking fumes pass through the purification system and the sampling is done on a silica cartridges impregnated with 2,4 DNPH (WATERS Sep-Pak DNPH-Silica). TO-11 method was adopted to collect and analyze carbonyls from cooking fume (US EPA) [15]. Teflon filter was set in front to remove particulate matter. The gas flowrate was 0.7 L/min, and the sampling time was for about one hour. An ozone scrubber was connected to the inlet of the 2,4-DNPH silica cartridge to prevent interference from ozone. All sampled cartridges were stored in laboratory refrigerators before analyzing 2,4-DNPH silica cartridge with high performance liquid chromatography (HPLC, Agilent 1260; Column: Zorbax RRHD Eclipse Plus C18 (2.1  $\times$  100 mm, 1.8  $\mu$ m)). Each 2,4-DNPH silica cartridge was eluted with 5.0 mL acetonitrile (HPLC grade) solution and transferred to a 2 mL volumetric flask before analyzing. The injection volume \was 18 µl. Mobile phase consisted of acetonitrile and water in the ratio 58%: 42%. The flow rate was 0.9 mL/min. The wavelength used was 365 nm and 430 nm.

Detailed information on the measurement procedure is contained in Table 1. Four factors were considered to be important for carbonyls emissions: cooking style (including barbecue, teppanyaki, frying and stir-frying); food materials (vegetables, meat and aquatic products) oil types (soybean oil, corn oil and sunflower oil) and the purification equipment (on/off). Control test was conducted by sampling carbonyls from the ambient air. In addition, the fuel we used was from the same batch with same quality, which were mixed evenly, and fully burned after 2 min, then the measurement started. For each cooking, we weighted all the ingredient and materials before the measurement. All the ingredient and



Fig. 1. Diagram of sampling methodology.

materials were taken from the same batch, respectively. And also the speed and frequency of the stir-frying process were controlled by a timer. Furthermore, we repeated three times in the same condition.

A high-voltage electrostatic hood (BF-JD-4, Shenzhen ENT Corporation, China Shenzhen, air volume  $= 20 \text{ m}^3/\text{min}$ ) were used for removing cooking emissions and cleaning room. It makes oil mist charged by the high-voltage electrode, and collected on the plate. Its purification efficiency is beyond 90% as claimed by the producer, and practical and no secondary pollution.

## 2.2. Emission factor calculation

Emission factors were calculated by multiplying the average concentration (C) by the cooking time (t), the flow from the metal container (F), the dilution ratio (DR), and dividing by the amount of food used  $(M_{Food})$  [16].

$$EF = \frac{C \times DR \times t \times F}{M_{Food}} \tag{1}$$

where,  $C (\mu g/m^3)$  is the average emission, DR is the dilution ratio, t (h) is the cooking time, F (m<sup>3</sup>/h) is flow rate,  $M_{Food}$  (kg) is the amount of food.

## 2.3. Health risk assessment and OFP

In this study, cancer risks were assessed by assuming inhalation of carbonyls produced by cooking emissions and calculated using US EPA (2011) guidelines as follows:

$$CR = slope factor \times LADD$$
 (2)

$$LADD = \frac{C \times IR \times AF \times ED}{BW \times AT}$$
(3)

where LADD (mg<sup>-1</sup> kg<sup>-1</sup> day<sup>-1</sup>) is the lifetime average daily dose,  $C(\text{mg/m}^3)$  is the pollutant concentration,  $IR(\text{m}^3/\text{day})$  is the intake rate, AF (%) is the absorption fraction, ED (day) is the exposure duration, BW(kg) is the body weight, and AT (day) is the average time days.

The values of input parameters were IR = 18 m<sup>3</sup>/day, AF = 100%, ED = 1.7 h × 24 h<sup>-1</sup> × 5 day/week × 52.14 week/year × 5 year = 92.34 day, BW = 64.56 kg, and AT = 27,959 days (=76.6 year) [17]. Total cooking time for three meals per day was 1.7 h in this study [17]. The slope factor in Eq. (2) was determined from reference dose (*RFD*, mg<sup>-1</sup> kg<sup>-1</sup> day<sup>-1</sup>) and according to the Integrated Risk Information System [18]. Note that only formaldehyde and acetaldehyde are carcinogenic among these substances. The CR value below 10<sup>-6</sup> has been set as to be acceptable or tolerable for regulating purposes [19].

The OFP of an individual carbonyl compounds can be calculated as multiplying the emission by its corresponding maximum increment reactivity (MIR) factor [20]. In this study, the total OFP is the summation of the OFPs corresponding to all carbonyl compounds.

#### 3. Results and discussion

#### 3.1. Carbonyls categories classified by carbon numbers

The emission factors of each carbonyl compound monitored in this study are given in Table 2.

The concentrations of C1-C3, in case of all four cooking styles, accounted for over 80% of the total carbonyls detected, while the content of C4-C8 was relatively low. Concentrations of chain-like

Tabla 1

Information of each measurement.

NO.	0. Cooking styles Fuel		Food materials	Oil types	Purification equipment	
1	Barbecue	Smokeless carbon		1	Electrostatic	
2	Barbecue	Smokeless carbon	Vegetables	Soybean oil	Electrostatic	
3	Barbecue	Smokeless carbon	Vegetables	Corn oil	Electrostatic	
4	Barbecue	Smokeless carbon	Vegetables	Sunflower oil	Electrostatic	
5	Barbecue	Smokeless carbon	Streaky pork	Soybean oil	Electrostatic	
6	Barbecue	Smokeless carbon	Squid-saury	Soybean oil	Electrostatic	
7	Barbecue	Smokeless carbon	Chicken-Chicken wing	Soybean oil	Electrostatic	
8	Barbecue	Smokeless carbon	Chicken wing	Soybean oil	Electrostatic + High energy particle	
9	Barbecue	Smokeless carbon	Chicken wing	Soybean oil	Electrostatic + High energy particle	
10	Barbecue	Smokeless carbon	Chicken wing	Soybean oil	No	
11	Barbecue	Smokeless carbon	Squid-saury	Soybean oil	No	
12	Teppanyaki	LPG	Vegetables	Soybean oil	Electrostatic	
13	Teppanyaki	LPG	Chicken -Chicken wing	Soybean oil	Electrostatic	
14	Teppanyaki	LPG	Tofu	Soybean oil	Electrostatic	
15	Teppanyaki	LPG	Streaky pork	Soybean oil	Electrostatic	
16	Teppanyaki	LPG	Pork	Soybean oil	No	
17	Frying	LPG	Pork	Soybean oil	No	
18	Frying	LPG	Patatoes	Soybean oil	No	
19	Stir-frying	LPG	Streaky pork	Soybean oil	No	
20	Stir-frying	LPG	Streaky pork	Soybean oil	Electrostatic + High energy particle	
21	Stir-frying	LPG	Streaky pork	Soybean oil	Electrostatic + High energy particle	
22	Stir-frying	LPG	Chicken	Soybean oil	Electrostatic + High energy particle	
23	Stir-frying	LPG	Chicken	Soybean oil	No	
24	Stir-frying	LPG	Chicken	Soybean oil	Electrostatic + High energy particle	
25	Stir-frying	LPG	Patatoes	Soybean oil	No	
26	Stir-frying	LPG	Patatoes	Corn oil	No	
27	Stir-frying	LPG	Patatoes	Sunflower oil	No	

C4-C8 carbonyls for both barbecue and teppanyaki were greater than that of cyclic C4-C8 carbonyls. Chain-like carbonyls recorded during frying and stir-frying were both lower than cyclic carbonyls. Feng et al. also concluded that higher content of fat and oil during the heating process resulted in higher emission of chain volatile compounds [21], The reason for this is the cooking temperature, which exhibited sudden increase in case of frying and stir-frying, and steady rise in case of barbecue and teppanyaki. A large amount of lower molecular weight aldehydes was generated due to the decomposition of fatty acids in food instead of oil. These were mainly chain-like carbonyls. Generally, barbeque and teppanyaki produced the greatest amount of carbonyls, frying produced less and stir-frying produced the least.

## 3.2. Impact of cooking style

Fig. 2 presents the content of five substances of carbonyl compounds for barbeque, teppanyaki, frying and stir-frying cooking styles. Aldehyde compounds were produced due to peroxyl radical reaction of fatty acid. Relatively high temperature was achieved during the cooking process to accelerate fatty acid cracking [22]. As a result, large amount of aldehydes were generated. Acroclein was formed from the dehydration of glycerol, and appeared among all types of edible oils tested. Acetone could have come from vegetables. Regarding the total EF of carbonyls, as shown in Table 2, different cooking styles showed the following trend: Barbecue > Teppanyaki > Frying > Stir-frying. Peng et al. [7] also found that, under the same other conditions, frying's emission concentrations were higher than for stir-frying.

Fig. 2 also shows that the primary pollutant was formaldehyde, followed by acetaldehyde and acetone. Acetaldehyde and propanal produced from barbecue were higher than in case of other cooking styles. Stir-frying produced very high content of formaldehyde and negligible amount of acetaldehyde produced. The total amount of carbonyls produced from teppanyaki cooking was lower than for barbecue. Overall, C1-C3 compounds accounted for more than 80%

Table 2

Categories of Carbonyl compounds and emission factors for each cooking style ( $\mu$ g/kg).

Compounds	Barbecue	Teppanyaki	Frying	Stir-frying
Formaldehyde	0.750 ± 0.106	$0.664 \pm 0.078$	0.747 ± 0.112	0.418 ± 0.127
Acetaldehyde	$0.384 \pm 0.089$	$0.051 \pm 0.011$	$0.204 \pm 0.057$	$0.000 \pm 0.000$
Propanal	$0.129 \pm 0.023$	$0.163 \pm 0.065$	$0.186 \pm 0.017$	$0.093 \pm 0.012$
Acetone	$0.158 \pm 0.022$	$0.179 \pm 0.034$	$0.269 \pm 0.065$	$0.132 \pm 0.075$
C1-C3	$1.421 \pm 0.325$	$1.057 \pm 0.321$	$1.407 \pm 0.354$	$0.643 \pm 0.145$
Butenal	$0.000 \pm 0.000$	$0.000 \pm 0.000$	$0.000 \pm 0.000$	$0.000 \pm 0.000$
Butyraldehyde	$0.068 \pm 0.004$	$0.024 \pm 0.008$	$0.000 \pm 0.000$	$0.005 \pm 0.002$
Pentanal	$0.047 \pm 0.010$	$0.043 \pm 0.007$	$0.000 \pm 0.000$	$0.000 \pm 0.000$
Hexanal	$0.028 \pm 0.004$	$0.014 \pm 0.003$	$0.000 \pm 0.000$	$0.000 \pm 0.000$
Isobutylene Aldehyde+2-butanone	$0.000 \pm 0.000$	$0.013 \pm 0.002$	$0.000 \pm 0.000$	$0.009 \pm 0.002$
Cyclohexanone	$0.010 \pm 0.003$	$0.062 \pm 0.011$	$0.116 \pm 0.032$	$0.038 \pm 0.006$
Benzaldehyde	$0.022 \pm 0.004$	$0.015 \pm 0.004$	$0.008 \pm 0.002$	$0.004 \pm 0.001$
2-Methylbenzaldehyde	$0.000 \pm 0.000$	$0.000 \pm 0.000$	$0.000 \pm 0.000$	$0.000 \pm 0.000$
m,p-Tolualdehyde	$0.000 \pm 0.000$	$0.000 \pm 0.000$	$0.000 \pm 0.000$	$0.000 \pm 0.000$
C4-C8 Chain	$0.144 \pm 0.056$	$0.095 \pm 0.034$	$0.000 \pm 0.000$	$0.014 \pm 0.006$
C4-C8 Cyclic annular	$0.032 \pm 0.008$	$0.077 \pm 0.005$	$0.123 \pm 0.064$	$0.042 \pm 0.003$
C4-C8	$0.175 \pm 0.064$	$0.172 \pm 0.039$	$0.123 \pm 0.064$	$0.056 \pm 0.009$
Total	$\textbf{1.596} \pm \textbf{0.389}$	$\textbf{1.229} \pm \textbf{0.360}$	$\textbf{1.530} \pm \textbf{0.418}$	$\textbf{0.699} \pm \textbf{0.154}$



Fig. 2. Impacts of cooking style on carbonyl compound substances.

of cooking fume for all cooking types. Formaldehyde contributed to 50% of the total carbonyls, followed by acetone and propanal. C4-C8 carbonyls content was found to be lower in this study. Other studies found that C1 -C3 carbonyls accounted for 51.46% of total carbonyl emissions during stir-frying with soybean oil. Formaldehyde was the highest, accounting for 17.77% [23]. Concentrations of C4-C8 carbonyls was also low. Ho et al. concluded that C1-C3 carbonyls accounted for 60.30% in barbeque emissions. In other research, formaldehyde was also the largest contributor, accounting for 25.43%, followed by acetaldehyde, nonanal, and acrolein [23]. Concentrations of nonanal produced from barbecue using soybean was the highest, accounting for 25.42%, followed by heptanal, octylaldehyde, formaldehyde, and acetaldehyde [11]. Hence formaldehyde was also one of the dominating carbonyl compounds, while reported concentrations of C4-C8 were low.

#### 3.3. Impact of food material

Type of food material has a great influence on the type of carbonyl compounds produced during cooking. Proteins, fat and other substances will decompose into smaller organic molecules after heating. It is known that the higher the amount of these materials, more organic pollutants will be generated accordingly. Also, it should be taken into consideration that this result may change due to different oil types, cooking types and different chemical reactions during cooking process. Main ingredients for each type of food used in this study are given in Table 3.

Fig. 3 presents emission factors of carbonyl compounds for different food materials used. Formaldehyde is dominating the emissions. Barbecue produced the greatest amount of carbonyls mainly due to the high content of fat. Carbonyls produced from pork were much higher than from other ingredient, mainly because high fatty acid content triggered the peroxyl radical reaction, which was the same for barbeque with chicken and seafood. Barbecuing vegetables produced less carbonyls [24]. Fuel used for barbeque and teppanyaki cooking were smokeless carbon and LPG, respectively. It is common for Chinese barbecue to use smokeless carbon as fuel, which is then continuously added during the cooking process to maintain a certain temperature. Studies have proved fuel combustion is also an important source of carbonyl compounds [3].

From Fig. 3 it can be seen that although fried meat and vegetables consume the same amount of oil, carbonyls produced from fried pork were lower than barbeque with pork. One possibility could be a higher temperature of the cooking oil, and the other reason could be relatively larger surface area of the oil layer due to the lack of stirring. The addition of molecular oxygen to triglyceride double bonds can occur\ on the oil surface. Carbonyls produced from stir-fried meat and vegetables were similar, which was much lower than for other food materials. Oil temperature, in this case, increased gradually, which lead to reduction of the formaldehyde. To get a better insight into these trends, we classified the food material into meat and non-meat types for each cooking type, as shown in Fig. 4.

From Fig. 4, it can be seen that formaldehyde has always been the most abundant carbonyl compound for each cooking type. Barbeque, teppanyaki and frying produced more carbonyls owning Cooking process was slow, so greater amount of lower aldehydes was emitted from the decomposition of fatty acids in meat than from oil [25]. Both frying and stir-frying started after the oil temperature rose to a certain value [16].

#### 3.4. Impact of edible oil

Impact of oil choice- in this case- soybean, corn and sunflower oil, on carbonyl emissions for barbecue and stir-frying cooking was investigated, was shown in Fig. 5. Generally, carbonyl emissions produced from soybean oil were higher than from corn oil. Sunflower seed oil was an exception, it emitted the least amount of carbonyl compound during stir-frying potato, but showed the greatest emissions for barbeque.

Table 3	
Main ingredient for each type of food (100 g) [31	J

Ingredient	Chicken	Pork	Cabbage	Tofu	Potato
Protein (g)	7.2	13.2	1.5	8.1	2
Fat (g)	16.4	37	0.1	3.7	0.2
Carbohydrate (g)	65.8	2.4	2.4	3.8	16.5
Ca (mg)	76	6	50	164	8
K (mg)	109	204	0	125	342
Cholesterol (mg)	81	80	0	0	0



Fig. 3. Impacts of food material on carbonyl compounds (dots represent the value of each sampling, columns notes the average values).

Fig. 5 also shows that cooking with soybean oil produces the highest amount of carbonyl compounds from stir-frying potato, but emissions are not too high from barbequing vegetables. Cooking with sunflower oil emitted the highest amount of carbonyls during barbequing vegetables, and the lowest from stir-frying potato. The main VOC detected was formaldehyde. C1-C3 were found to be the major contributor. In the process of stir-frying potatoes with sunflower oil, methacrolein + 2-butanone were produced in a large amount. Comparatively, cooking with other oils did not produce these species. The total amount of carbonyls was significantly high during stir-frying potatoes using LPG, which was possibly due to the presence of large amounts of water in materials [26]. Sunflower oil was unsuitable for barbecues as it was found that e large amounts of acetaldehyde were formed from unsaturated fatty acids during the heating process. Emissions of the corn oil were smaller, attributing to the low amount of linolenic acid (31.66%) in corn oil. The autoxidation rate of linolenic acid is 77 times higher than that of oleic acid [27].

Table SI 1 shows the main components of the three types of oil, which were oleic and linoleic acid. Metabolites and volatile substances decomposed by long chain hydrocarbon (alkene) species will escape from crude oil after heating. In addition, linoleic acids have more double-bonds, so it can produce more of lower carbonyl compounds.

## 3.5. Impact of purification system

High-voltage electrostatic methodology is always used for cooking fume purification, mainly for particulate matters. Fig. 6 shows emission factors of three cooking types before and after purification, among of which, fried pork showed no significant difference. Carbonyls produced from fried chicken were reduced by 50%. Unexpectedly, carbonyls emitted from roast chicken wings were even greater than before purification.

The working principle of high-voltage electrostatic precipitator relies on the charging of particulate matter produced from cooking, and transport of PM towards to polar plate under high voltage [28]. Discharge process of high-voltage electrostatic precipitator can cause a portion of organic matters with high molecular weight to decompose into organic compounds with low molecular weight, such as carbonyls. In addition, ozone could be produced during the discharge process as well as the high-energy particle deodorization device. Ozone further oxidizes organic hydrocarbons into carbonyl compounds.

#### 3.6. OFP

The concept of OFP is used widely to assess the roles of ambient VOC species in ozone formation [29]. The OFP contributions of carbonyl compounds are shown in Fig. 7.

Fig. 7 indicates that species with greater concentrations may not be the major contributor to OFP, such as acetone and C4-C8 carbonyl compounds. Owning to higher MIR of formaldehyde, formaldehyde not only dominated the carbonyls emissions, accounting for 47.0–60.0%, but also contributed the most to OFP, reaching up to 56.1–79.9%.

## 3.7. Health risk assessment

Carcinogenic compounds, such as formaldehyde and acetaldehyde, were produced with large amount from residential cooking activities as discussed previously [30]. According to Eqs (2) and (3), cancer risks (CR) related to the exposure to carbonyls are investigated for four cooking styles and three oil types, as given by Tables 4 and 5.



Fig. 4. Impacts of meat and non-meat food material on carbonyl compounds for each cooking type (dots represent the value of each sampling, columns notes the average values).



Fig. 5. Impacts of edible oil on carbonyl compounds produced from stir-frying potato and barbeque vegetable.



Fig. 6. Impacts of purification equipment on removing carbonyl compound.



Formaldehyde XXX Acetaldehyde XXX Propanal Acetone H C4-C8

Fig. 7. Carbonyl compounds contributions to OFP for different cooking styles.

The cancer risk ranged between  $1.51 \times 10^{-7}$  and  $2.66 \times 10^{-7}$  for exposure to carbonyls produced from different cooking styles, all of which were below the limit of  $10^{-6}$ . The limit value for formaldehyde regulated by China standard is  $0.10 \text{ mg/m}^3$ , the measured concentration of  $2.16-19.70\mu\text{g/m}^3$  is lower than the standard. Frying contributed the most due to high emission of formaldehyde (Table 2). For oil types, adoption of sunflower oil for barbeque vegetable and soybean oil for stir-frying potato were with associated with high possibilities for cancer risk, because emissions from sunflower oil and soybean oil were the highest for barbeque vegetable and stir-frying potato, respectively.

# Table 4

Cancer risk (CR) assessments of exposing to carbonyl compounds for different cooking styles.

Cooking style	Cancer risk (CR)
Barbecues	2.35E-07
Teppanyaki	2.08E-07
Frying	2.66E-07
Stir-frying	1.51E-07

#### Table 5

Cancer risk (CR) assessments of exposing to carbonyl compounds for different oil types.

	Barbeque vegetable			Stir-frying potato		
	Soybean oil	Corn Oil	Sunflower Oil	Soybean oil	Corn Oil	Sunflower Oil
Cancer risk (CR)	5.65E-08	6.62E-08	1.30E-07	1.29E-07	1.06E-07	4.17E-08

## 4. Conclusion

The characteristics of carbonyl emissions produced from cooking have been investigated, considering factors including cooking styles, oil types, food materials, and purification facilities. C1-C3 carbonyl compounds accounted for more than 80% of the total carbonyls. Among all factors, the major pollutants were formaldehyde, acetaldehyde and acetone. Emission trend for different cooking styles was in a following order: barbecue > frying > teppanyaki > stir-frying, and for food materials: meat dishes > non-meat dishes. Cooking with sunflower oil produces the highest concentrations of carbonyls, especially for barbecue cooking. High voltage electrostatic precipitation had no obvious effect on removing carbonyls. Moreover, assessments on human health effects indicate that frying carries the most serious potential for health risk, mainly due to the high concentrations of formaldehyde, which was also the reason for the highest OFP contribution.

Cooking fume pollution affects indoor air quality significantly and is associated with negative health effects. People exposed to cooking emissions on a regular basis should pay attention to keep the flue smooth and take appropriate self-protection measures; At the same time, we should further advance out understanding of cooking fumes most of the chefs and stay-at-home women are exposed to, and use advanced technical means to remove carcinogens from fumes Future experiments should focus more on carcinogens, and examine PM concentrations and particle size distribution during the tests. New standards are also necessary for both air purification system and indoor air quality.

#### Notes

The authors declare no competing financial interest.

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# Appendix A. Supplementary data

Supplementary data related to this article can be found at http://dx.doi.org/10.1016/j.buildenv.2017.08.045.

#### References

- J.Y. Zheng, M. Shao, W.W. Che, Speciated VOC emission inventory and spatial patterns of ozone formation potential in the Pearl River Delta, China, Environ. Sci. Technol. 43 (2009) 8580–8586.
- [2] J.C. Cheng, T. Cui, W.Q. He, Pollution characteristics of aldehydes and ketones compounds in the exhaust of Beijing typical restaurants, Environ. Sci. 36 (2015) 2743–2748.
- [3] X.H. Li, S.X. Wang, J.M. Hao, Characteristics of volatile organic compounds (VOCs) emitted from biofuel combustion in China, Chin. J. Environ. Sci. 32 (2011) 3515–3521.
- [4] M.T. Wen, M. Hu, Physical and chemical characteristics of fine particles emitted from cooking emissions and its contribution to particulate organic

matter in Beijing, Chin. J. Environ. Sci. 28 (2009) 2620-2625.

- [5] J. Zhang, P.J. Lioy, Q. He, Characteristics of aldehydes: concentrations, sources, and exposures for indoor and outdoor residential microenvironments, Environ. Sci. Technol. 28 (1994) 146–152.
- [6] S.H.H. Steven, Z.Y. Jian, W.C. Kam, Carbonyl emissions from commercial cooking sources in Hong Kong, J. Air Waste Manag. Assoc. 56 (2006) 1091–1098.
- [7] C.Y. Peng, C.H. Lan, P.C. Lin, Y.C. Kuo, Effects of cooking method, cooking oil, and food type on aldehyde emissions in cooking oil fumes, J. Hazard. Mater. 324 (2017) 160.
- [8] B.Y. Zhang, Y.Z. Yang, Discussion on the composition, harm and preventing and controlling methodes of the cooking oil fumes, Sichuan Food And Ferment. 42 (2006) 14–18.
- [9] Y.L. Feng, A Preliminary Study of Carbonyl Compounds and Their Sources of Indoor and Outdoor Air in Guangzhou (In Chinese), Chinese Academy of Sciences, 2005.
- [10] Y.X. Yu, Carbon Isotopes of Atmospheric Carbonyl Compounds and Atmospheric Organic Pollutants (In Chinese), Guangzhou Institute of Geochemistry, 2006.
- [11] S.S. Ho, J.Z. Yu, K.W. Chu, Carbonyl emissions from commercial cooking sources in Hong Kong, J. Air & Waste Manag. Assoc. 56 (2006) 1091–1098.
- [12] C.Z. Shi, X. Jiang, Z.L. Yao, J. Li, X.W. Hao, Y. Yin, et al., Carbonyl compounds emission characteristics in cooking fumes, Chin. J. Environ. Eng. 9 (2015) 1376–1380.
- [13] X. Zhu, K. Wang, J. Zhu, Analysis of cooking oil fumes by ultraviolet spectrometry and gas chromatography-mass spectrometry, J. Agric. Food Chem. 49 (2001) 4790–4794.
- [14] H. Guo, T. Wang, D.R. Blake, I.J. Simpson, Y.H. Kwok, Y.S. Li, Regional and local contributions to ambient non-methane volatile organic compounds at a polluted rural/coastal site in Pearl River Delta, China, Atmos. Environ. 40 (2006) 2345–2359.
- [15] Norma T. Murphy, Compendium of Methods for the Determination of Toxic Organic Compounds in Ambient Air, Atmospheric Research and Exposure Assessment Laboratory, US Environmental Protection Agency, 1989.
- [16] F. Klein, S.M. Platt, N.J. Farren, A. Detournay, E.A. Bruns, C. Bozzetti, Characterization of gas-phase organics using proton transfer reaction time-of-flight mass spectrometry: cooking emissions, Environ. Sci. Technol. 50 (2016) 1–35.
- [17] J.H. Cheng, Y.S. Lee, K.S. Chen, Carbonyl compounds in dining areas, kitchens and exhaust streams in restaurants with varying cooking methods in Kaohsiung, Taiwan, J. Environ. Sci. 41 (2016) 218–226.
- [18] US-EPA, Integrated Risk Information System (IRIS), 2015.
- [19] X. Hu, Y. Zhang, Z. Ding, T. Wang, H. Lian, Y. Sun, Bioaccessibility and health risk of arsenic and heavy metals (cd, co, cr, cu, ni, pb, zn and mn) in tsp and pm2.5 in Nanjing, China, Atmos. Environ. 57 (2012) 146–152.
- [20] W. Carter, Saprc Atmospheric Chemical Mechanisms and Voc Reactivity Scales, 2010.
- [21] Y.L. Feng, J. Huang, S. Wen, Concentration and pattern of carbonyl compounds from oil smoke in restaurant, Environ. Sci. Technol. 31 (2008) 66–68.
- [22] J.J. Schauer, M.J. Kleeman, G.R. Cass, B.R. Simoneit, Measurement of emissions from air pollution sources. 4. C1-C27 organic compounds from cooking with seed oils, Environ. Sci. Technol. 36 (2002) 567.
- [23] Y. Zhang, Y. Mu, J. Liu, Levels, sources and health risks of carbonyls and BTEX in the ambient air of Beijing, China, J. Environ. Sci. 24 (2012) 124–130.
- [24] J. James, Michael J. Schauer, Glen R. Kleeman, Cass, et al., Measurement of emissions from air pollution sources. 1. c1 through c29 organic compounds from meat charbroiling, Environscitechnol 33 (1999) 1566–1577.
- [25] J.D. Wood, R.I. Richardson, G.R. Nute, A.V. Fisher, M.M. Campo, E. Kasapidou, et al., Effects of fatty acids on meat quality: a review, Meat Sci. 66 (2004) 21–32.
- [26] K.L. Abdullahi, J.M. Delgado-Saborit, R.M. Harrison, Emissions and indoor concentrations of particulate matter and its specific chemical components from cooking: a review, Atmos. Environ. 71 (2013) 260–294.
- [27] C. Izard, C. Liberman, Acrolein, Mutat. Research/Reviews Genet. Toxicol. 47 (1978) 115–138.
- [28] L.Y. He, M. Hu, X.F. Huang, Measurement of emissions of fine particulate organic matter from Chinese cooking, Atmos. Environ. 3 (2004) 6557–6564.
- [29] W.P.L. Carter, Development of ozone reactivity scales for volatile organic compounds, J. Air Waste Manage Assoc. 44 (1994) 881–899.
- [30] Y. Huang, S.S.H. Ho, K.F. Ho, Characteristics and health impacts of VOCs and carbonyls associated with residential cooking activities in Hong Kong, J. Hazard. Mater. 186 (2011) 344–351.
- [31] J.T. Zhang, Z.Y. Wang, Food and Nutrition(In Chinese), Shanghai Scientific & Technical Publishers, 1983.