

1 We thank the reviewers for their careful review of the manuscript. The comments greatly improved  
2 our manuscript. We revised our manuscript according to the reviewers' comments and suggestions.  
3 Overall, we have changed the mass concentration ( $\mu\text{g m}^{-3}$ ) to the emission rate ( $\mu\text{g min}^{-1}$ ) to avoid  
4 the influence of cooking time and sampling time according to the comments of the referees. We add  
5 more details to the volatility distributions of cooking emissions. We also added more comparisons  
6 with different studies. Following are our responses to the comments.

7

8 **Response to referee #3:**

9 This manuscript investigates the impact of cooking style and oil on the emissions from traditional  
10 Chinese cooking. A significant number of chemical species including aromatics, alkanes, oxygenated  
11 compounds, and PAHs have been detected. The authors observed that in addition to VOC species,  
12 S/IVOCs made up an important fraction of cooking emissions and SOA precursors. In general, dishes  
13 cooked by stir-frying and deep-frying styles emit more pollutants than relatively mild cooking styles.  
14 A volatility-polarity distribution framework of cooking emissions has been developed. Unlike the  
15 emissions that showed great variation, the volatility-polarity distribution of different cooking styles  
16 was similar. PLS-DA and MPCA analyses revealed that cooking oil was a critical influencing factor  
17 in the 2D distribution. Overall, this is a comprehensive study investigating the relationship among  
18 cooking emissions, cooking styles, and cooking materials. The manuscript is well written, and the  
19 results are valuable to the literature. I would like to recommend its publication in Atmospheric  
20 Chemistry and Physics, subject to minor revisions.

21 Thank you for your comments. The valuable suggestions addressed have greatly improved our  
22 manuscript. Following are our point-to-point responses to the comments.

23

24 1. Table S1: In regard to oil temperature, how was oil temperature measured and monitored? Was  
25 oil temperature controlled and maintained the same during the cooking? There seems to be a positive  
26 relationship between oil temperature (Table S1) and emissions (Figure S3). Have the authors tried to  
27 cook the dishes at the same oil temperature and compare the emission results?

28 Thank you for your comment. The oil temperature was measured by a thermometer placed in the oil.

29 The thermometer was removed from the oil before placing the cooking materials. As a result, the  
30 *initial* temperature of the oil was maintained the same for each dish. Dishes cooked at the same oil  
31 temperature were not conducted in this work. Further investigation will be carried on to illustrate the  
32 relationship between oil temperature and cooking emissions.

33 We revised Table S1 as follows.

34 **Table S1.** Details of cooking procedures.

<b>Domestic cooking</b>	<b>Material</b>	<b>Oil temperature<sup>#</sup></b>
Fried chicken	170 g chicken, 500 mL oil (corn, peanut, soybean, or sunflower oil), a few condiments	145 ~ 150 °C
Kung Pao chicken	150 g chicken, 50 g peanut, 40 mL corn oil, a few condiments	Not stable
Pan-fried tofu	500 g tofu, 200 mL corn oil, a few condiments	100 ~ 110 °C
Stir-fried cabbage	300 g chicken, 40 mL corn oil, a few condiments	95 ~ 105 °C

35 <sup>#</sup> The oil temperature was measured by a thermometer placed in the oil. The thermometer was  
36 removed from the oil before placing the cooking materials. The temperatures listed in Table S1 were  
37 initial cooking temperatures and were maintained the same for each dish.

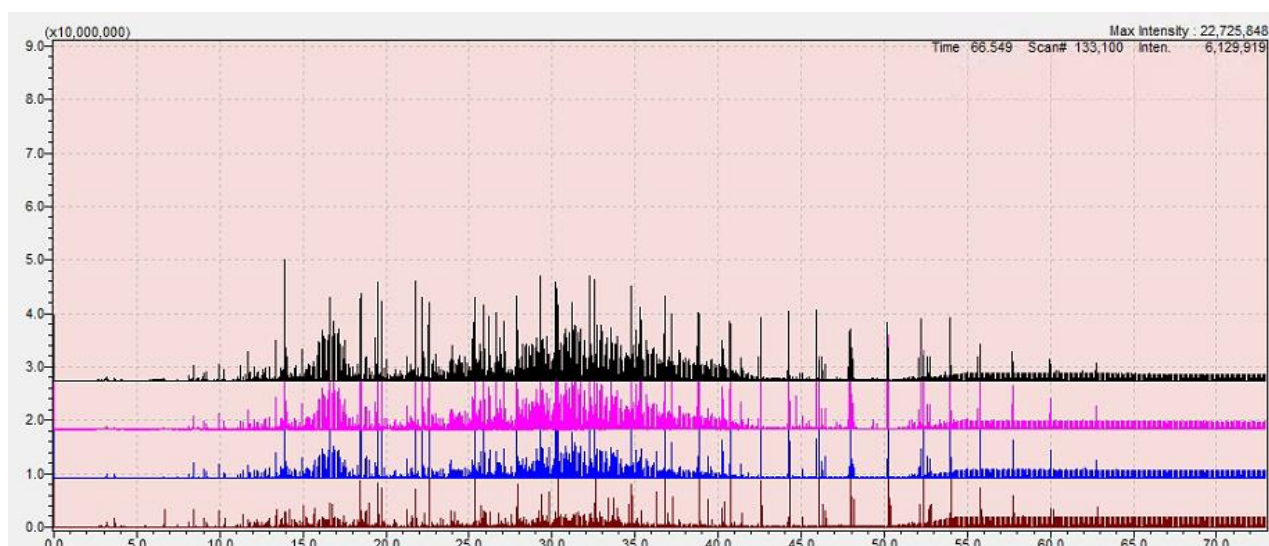
38

39 2. Line 117: What's the dimension of the Tenax TA tube? A flow rate of 0.5 L min<sup>-1</sup> was used in  
40 this study. Do you have any idea what were the collection efficiencies of chemical species with  
41 different volatility under this flow rate condition? How long was the sampling? What about the  
42 breakthrough of Tenax TA tubes?

43 Thank you for your comment. The Tenax TA tube is Gerstel 6 mm 97 OD, 4.5 mm ID glass  
44 tube filled with ~290 mg Tenax TA. A Tenax TA breakthrough experiment was conducted by  
45 introducing pure nitrogen gas (N<sub>2</sub>) with a flow of 0.5 L min<sup>-1</sup> to the desorption tube with pre-added  
46 standard chemicals (Figure S2). No significant breakthrough was observed within 24 h (<3% of TIC).

47 The sampling time in this work is 15 ~ 30 min ( $0.5 \text{ L min}^{-1}$ ) which is much less than 24h. The  
48 chemical species quantified in this work was stable on Tenax TA tubes even after 24h of  $\text{N}_2$  flowing.  
49 We have revised the manuscript accordingly.

50 Cooking fumes were sampled directly without dilution. After collecting particles on quartz filters,  
51 gas-phase organics were sampled by pre-conditioned Tenax TA tubes (Gerstel 6 mm 97 OD, 4.5  
52 mm ID glass tube filled with ~290 mg Tenax TA) with a flow of  $0.5 \text{ L min}^{-1}$ . The  
53 removal of particles on the quartz filter in front of the Tenax TA tubes affects the S/IVOC  
54 measurements, causing positive and negative artifacts. Some of the gaseous SVOCs could be lost to  
55 sorption onto filters, and some particle-phase SVOCs could evaporate off the filter. The emission  
56 pattern of the particulate organics diverged from gas-phase organics, and a small overlap of species is  
57 identified. Aromatics, aldehydes, and short-chain acids mainly occurred in the gas-phase. For  
58 instance, the detection of short-chain olefinic aldehydes in the gas-phase was 40 times that of the  
59 particle-phase aldehydes. The artifacts of particulates on gas-phase aromatics and oxygenated  
60 compounds could be less than 5%. A typical system blank chromatogram is displayed in Figure S1. A  
61 daily blank sampling of the air in the kitchen ventilator was conducted before cooking and was  
62 subtracted in the quantification procedure. All samples were frozen at  $-20^\circ\text{C}$  before analyzing. A  
63 Tenax TA breakthrough experiment was conducted by introducing pure nitrogen gas ( $\text{N}_2$ ) with a flow  
64 of  $0.5 \text{ L min}^{-1}$  to the desorption tube with pre-added standard chemicals (Figure S2). No significant  
65 breakthrough was observed within 24 h (<3% of TIC). The sampling time in this work is 15 ~ 30 min  
66 ( $0.5 \text{ L min}^{-1}$ ) which is much less than 24h.



67

68 Figure S2. The chromatograms of standard chemicals after 6h (brown), 24h (blue), 48h (red), and  
 69 72h (blue) of flowing by pure nitrogen gas. The flow of nitrogen gas is set to be the same as the  
 70 sampling flow ( $0.5 \text{ L min}^{-1}$ ). No significant breakthrough was observed within 24 h (<3%).

71

72 3. Lines 120-131: Chemical analysis using TD may have the following concerns (taking SVOCs  
 73 as examples):

74 a) Some of the SVOCs are of relatively low volatility. A TD temperature of  $280 \text{ }^\circ\text{C}$  may not be  
 75 sufficient to thermally released all the SVOCs in a short period of time.

76 b) SVOCs such as acids may get decomposed during the TD processes.

77 c) The decomposition of SVOCs may produce small molecules that can be mistakenly  
 78 identified as VOCs.

79 Both items a and b lead to underestimations of SVOCs. Item c may result in an overestimation of  
 80 VOCs. In regard to these concerns, how long was the TD process in this study? Have the authors  
 81 quantified the desorption efficiency of SVOC standards?

82 Thank you for your comment. The programming of the TD process was ramped  $30^\circ\text{C}$  to  $280^\circ\text{C}$   
 83 ( $60^\circ\text{C}/\text{min}$ ) and then retained at  $280^\circ\text{C}$  for 10 min (Table S2). The total thermal desorption time was  
 84 14 min.  $280^\circ\text{C}$  was chosen for thermal desorption temperature due to the less bleeding of Tenax TA  
 85 compared with  $300^\circ\text{C}$ . The linearities of undecanoic acid (C11-acid), C31, and C32 were 0.97, 0.99,  
 86 and 0.99 (Table S5). The good linearity of SVOC compounds under different concentration levels

87 showed a good desorption efficiency of SVOCs. Furthermore, the desorption of SVOC occurred in  
88 both *standards* and *samples*, making the quantification face less uncertainty. Though the direct  
89 desorption efficiency of SVOC is not quantified, we add more uncertainty discussions to the  
90 implication part of the manuscript as follows.

91 We still need to stress that although GC×GC is utilized, UCMs still occur sharing a proportion of  
92 5% of the total response in this work. Acids and aldehydes tail in the second column and cause  
93 uncertainties in the quantification procedure. Meanwhile, TD-GC×GC-MS does not  
94 comprehensively measure all compounds. Acids can decompose during thermal desorption if no  
95 derivatization was performed. Meanwhile, the decomposition of SVOC compounds could produce  
96 small molecules in the VOC or IVOC range. The TD process could introduce underestimation for  
97 SVOC compounds while causing overestimations of VOC and IVOC species. Highly polar  
98 compounds do not elute from the GC column. This may lead to biases in estimating volatility and  
99 polarity distributions. Comparisons between GC×GC and chemical ionization mass spectrometers  
100 (CIMS) should be further implemented to give a full glimpse of cooking organic compounds.

101

102 4. Line 126: The authors mentioned that the chromatogram was cut into different volatility bins  
103 (B9 to B31 with a decrease in volatility). However, Figure 2 and Table S3 start from “B8\_before”.  
104 Please clarify.

105 Please add a sentence in the text defining the volatility of each bin (e.g., B8). Please also add a  
106 sentence in the text defining the polarity of each bin (e.g., P1). In this way, other studies can compare  
107 their results to this study when the volatility-polarity distribution framework is used.

108 Thank you for your comment. We have changed the statement of B9 to B8 as the 1D bins started  
109 with B8\_before. We add instances of C12 and benzophenone to the main text to further illustrate the  
110 2D binning method.

111 The total chromatogram was cut into volatility bins (B8 to B31 with a decrease in volatility)  
112 following the pipeline of previous studies (Tang et al., 2021; Zhao et al., 2014, 2017, 2018), while it  
113 was cut into slices by an increase of 0.5 s in the second retention time (called 2D bins, from P1 to  
114 P12 with an increase of polarity). For instance, C12 lies in B12 (saturated vapor concentration  $\sim 10^6$

115  $\mu\text{g m}^{-3}$ , IVOC range) and P2 bins (low polarity). Benzophenone lies in B16 (saturated vapor  
116 concentration  $\sim 10^5 \mu\text{g m}^{-3}$ , IVOC range) and P6 bins (medium to high polarity). A two-dimensional  
117 panel was developed in this way to investigate the emission of contaminants from aspects of their  
118 volatility and polarity properties (Song et al., 2022).

119

120 5. Equation 2: SOA yield of VOC can increase with increasing particle loading (Odum et al.,  
121 ES&T, 1996). Were the values of SOA yields used herein the maximum SOA yields? Please clarify.

122 Thank you for your comment. The SOA yields utilized in this work are under high  $\text{NO}_x$  conditions  
123 which are underestimation of SOA due to the lower yields compared to low  $\text{NO}_x$  conditions. We have  
124 revised the manuscript as follows.

125 SOA ( $\mu\text{g min}^{-1}$ ) was estimated by the following equation, where  $[\text{HC}_i]$  is the emission rate of  
126 precursor  $i$  ( $\mu\text{g min}^{-1}$ ) with OH reaction rate of  $k_{\text{OH},i}$ , ( $\text{cm}^3 \text{ molecules}^{-1} \text{ s}^{-1}$ ) and SOA yield of  $Y_i$   
127 (Table S3). The SOA yields of precursors were from literature (Algrim and Ziemann, 2016, 2019;  
128 Chan et al., 2009, 2010; Harvey and Petrucci, 2015; Li et al., 2016; Liu et al., 2018; Loza et al., 2014;  
129 Matsunaga et al., 2009; McDonald et al., 2018; Shah et al., 2020; Tkacik et al., 2012; Wu et al., 2017)  
130 or surrogates from  $n$ -alkanes in the same volatility bins (Zhao et al., 2014, 2017). The SOA yields  
131 utilized in this work are under high  $\text{NO}_x$  conditions which are underestimation of SOA due to the  
132 lower yields compared to low  $\text{NO}_x$  conditions.  $[\text{OH}] \times \Delta t$  is the OH exposure and was set to be  
133  $14.4 \times 10^{10} \text{ molecules cm}^{-3} \text{ s}$  ( $\sim 1.1$  days in OH concentration of  $1.5 \times 10^6 \text{ molecules cm}^{-3}$ ) in order to  
134 keep pace with our previous work (Zhang et al., 2021b; Zhu et al., 2021).

$$135 \quad \text{SOA} = \sum[\text{HC}_i] \times (1 - e^{-k_{\text{OH},i} \times [\text{OH}] \times \Delta t}) \times Y_i \quad (3)$$

136

137 6. Lines 220-222: The authors mentioned that “an enhancement of ozone formation contribution  
138 and a decrease of SOA formation contribution were observed”. The sentence is confusing. In regard  
139 to “enhancement” and “decrease”, what were you comparing? Different types of VOCs, or VOCs vs.  
140 S/IVOCs, or VOC emissions from different cooking styles?

141 Thank you for your comment. We compared the contribution to the mass proportion of VOCs in ERs.

142 We have revised the manuscript as follows.

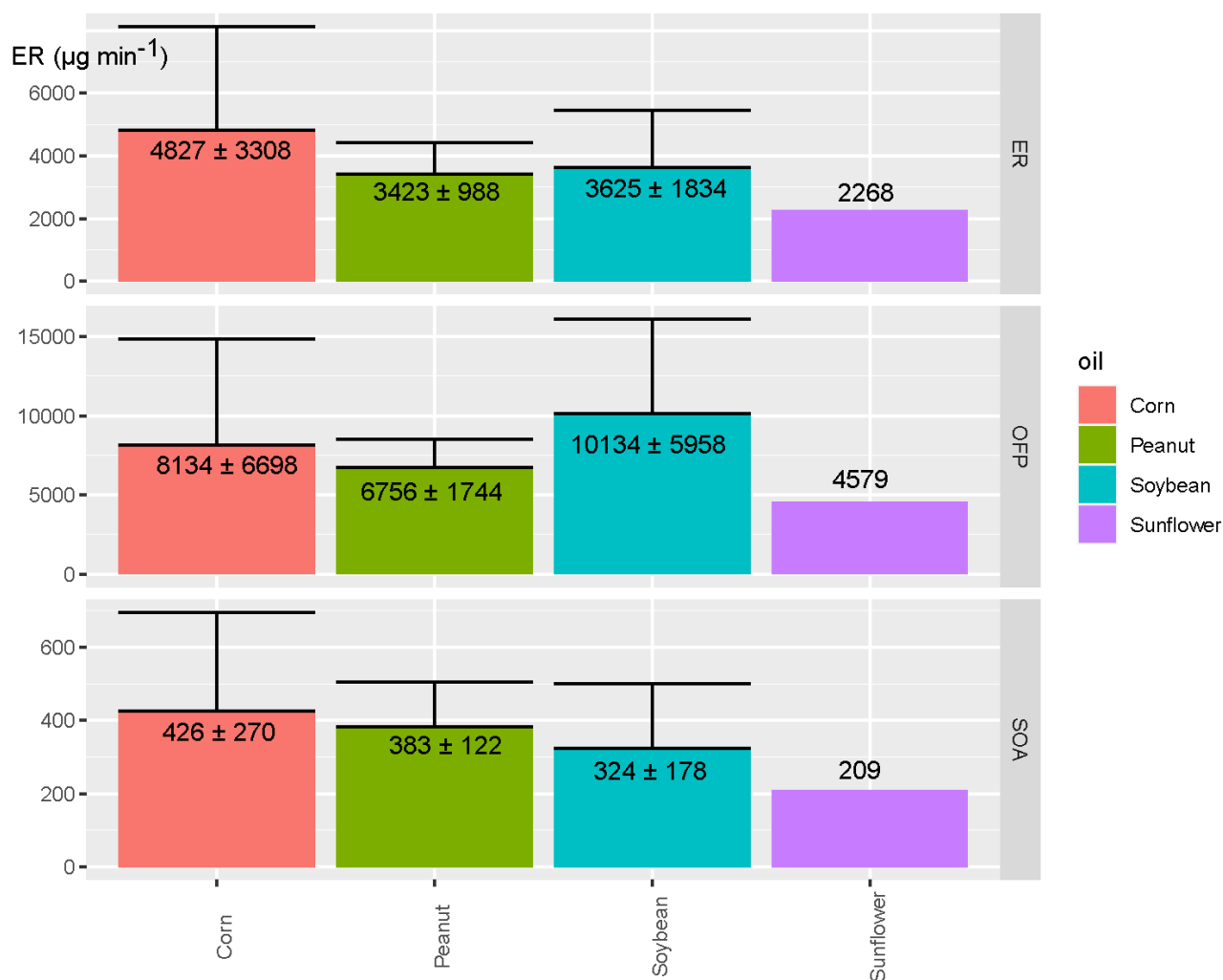
143 Although chemicals in the VOC range dominated ozone and SOA formation, an increase in ozone  
144 formation contribution and a decrease in SOA formation contribution compared with the mass  
145 proportion of VOCs in ERs were observed. VOCs contributed 90.3% - 99.8% of the ozone  
146 estimation, and 68.0% - 89.8% of the total SOA estimation, compared with 81.4% - 95.6% in ERs.  
147 S/IVOCs explained 10.2% - 32.0% of the SOA estimation.

148

149 7. Lines 236-237: The authors mentioned that “the emission patterns diverged from heated oil  
150 fumes as heated sunflower oil and peanut oil emitted more organics”. It seems that this statement  
151 conflicts with the results shown in Figure S7 (dishes cooked by sunflower oil had the lowest  
152 emission).

153 Thank you for your comment. We have revised the manuscript as follows.

154 Chicken fried with corn oil emitted the most abundant gaseous contaminants. The emission patterns  
155 in this work diverged from heated oil fumes (Liu et al., 2018) as in their work heated sunflower oil  
156 and peanut oil emitted more organics.



157

158 Figure 4. Emission rate (ER), ozone formation potential (OFP), and secondary organic aerosol (SOA)  
 159 estimation from emissions of fried chicken cooked with corn, peanut, soybean, and sunflower oils.

160 The unit of the y-axis is  $\mu\text{g min}^{-1}$ .

161

162 8. Lines 265-266: “In contrast, the volatility-polarity distributions of dishes did not vary much  
 163 when corn oil was used for cooking”. Please add a reference to Figure 2.

164 Thank you for your comment. We have deleted this statement. The revised manuscript is shown as  
 165 follows.

166 Although pollutants were dominated by aromatics, alkanes, and oxygenated compounds with  
 167 volatility bins of B9 to B12 (VOC-IVOC range, saturated vapor concentration  $> 10^6 \mu\text{g m}^{-3}$ ) and  
 168 polarity bins of P1 to P5 (low to medium polarity), significant diversities of volatility-polarity  
 169 distributions were observed (Figure S9). The chemical compositions in each volatility bin were also



170 distinct (Figure S11). IVOCs accounted for as much as 22.8% and 23.7% of the total ERs when  
171 peanut and sunflower oils were utilized for frying (Kostik et al., 2013; Ryan et al., 2008). The peanut  
172 oil was much more abundant in oleic acid (41.5%), while the proportion of linoleic acid in sunflower  
173 is 36.6% (Figure S10). The proportion of unsaturated acids in peanut and sunflower oils is higher  
174 than that of other oils.

175

176 9. Line 278: SOA production or reduction?

177 Thank you for your comment. We revised the manuscript as follows.

178 Despite the importance of aldehydes revealed in previous studies (Klein et al., 2016; Liu et al., 2018),  
179 our results demonstrated that alkanes, pinenes, and short-chain acids are also key precursors in  
180 cooking SOA **production** (Huang et al., 2020).

181

182 10. Lines 294-295: What do you mean by “physical reactions (evaporation)”? Evaporation of  
183 what?

184 Thank you for your comment. We revised the manuscript as follows.

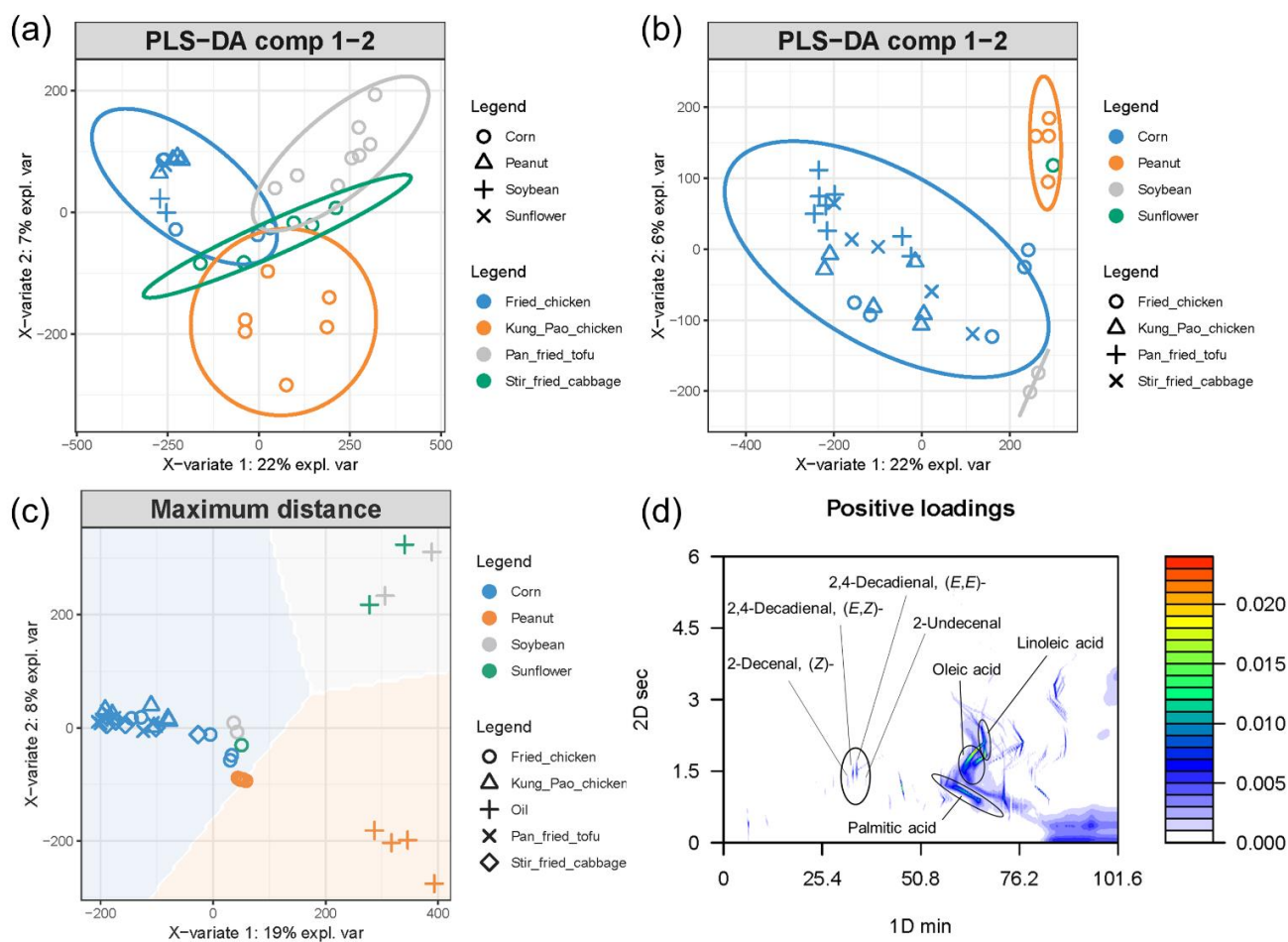
185 The PLS-DA result showed that cooking emissions diverged from oils (Figure 5 (c)), indicating that  
186 the physical reactions (**evaporation of edible oils**) were not the main reactions during the cooking  
187 procedure.

188

189 11. Lines 295-296: “MPCA results showed the chromatogram similarities (positive loading) of oils  
190 and emissions.” Please add a reference to Figure 3d. What is the color bar of Figure 3d?

191 Thank you for your comment. We add a reference to Figure 5d. The color bar in Figure 5(d) is the  
192 positive loading of pixels. We revised the manuscript as follows.

193 MPCA results showed the chromatogram similarities (positive loading) of oils and emissions  
194 (**Figure 5(d)**).



195

196 **Figure 5. PLS-DA classification results in setting the cooking style (a) or oil (b) as grouping**  
 197 **variables. When oil was set as the grouping variable, the separation of groups was much better than**  
 198 **setting the dish as the grouping variable. The PLS-DA comparison result of cooking emissions and**  
 199 **oils is displayed in (c), indicating that the cooking fume is not just the evaporation of oil itself.**  
 200 **Positive loadings of oil and cooking fume chromatograms (d) demonstrated the key components**  
 201 **contributing to the similarities of samples. The color bar in (d) is the positive loading of pixels.**

202

203 Technical comments:

204 1. Line 167: duplicate word “form”

205 Thank you for your comment. We revised the manuscript as follows.

206 Chromatograms were imported from the network common data form (netCDF).

207

208 2. Line 174: Change “results” to “result”

209 Thank you for your comment. We revised the manuscript as follows.

210 PLS-DA is a supervised method for the classification of grouped data. The main influencing factor  
211 could be apportioned if one separation **result** of PLS-DA is much better than the other.

212

213 3. Line 313: Change “gas-phase” to “gas phase”

214 Thank you for your comment. We revised the manuscript as follows.

215 These highly volatile contaminants escape from oil immediately and lead to an accumulation of  
216 oxygenated compounds in the **gas phase**.

217

218 **Reference:**

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