We thank the reviewers for their careful review of the manuscript. The comments greatly improved our manuscript. We revised our manuscript according to the reviewers' comments and suggestions. Overall, we have changed the mass concentration (µg m⁻³) to the emission rate (µg min⁻¹) to avoid the influence of cooking time and sampling time according to the comments of the referees. We add more details to the volatility distributions of cooking emissions. We also added more comparisons with different studies. Following are our responses to the comments.

7

8 **Response to referee #1:**

9 In this manuscript, the authors studied the gaseous emissions from Chinese domestic cooking and the 10 impact of cooking style and oil used on the emission profiles. They performed cooking experiments 11 at a laboratory facility and measured the detailed composition of gaseous compounds using 12 multidimensional GC-MS. They observed that the oil type played the most important role in determining the volatility and polarity distribution of compounds, while the type of food cooked and 13 14 cooking style influenced the detailed composition, but was less of a factor in determining overall 15 volatility and polarity. They also highlighted the role of IVOCs and SVOCs, which are not as well 16 measured in previous studies but can add 10-30% to estimated SOA formation. All of these 17 observations are important for understanding food cooking emissions as a source of reactive organic 18 compounds in the urban atmosphere. The experiments are well-designed and the results are 19 thoroughly interpreted and explained. The manuscript is often difficult to understand so I would 20 recommend major revisions, mostly for the sake of improving the clarity of the manuscript. 21 Otherwise the technical content is suitable for publication in ACP.

We greatly thank the reviewer for his careful review of the manuscript. Following are ourpoint-to-point responses to the comments.

24

25 Major comments:

All of the emissions are reported in air concentrations (ug per m3 of air sampled). These numbers would depend on air flow rate through the cooking apparatus, which may vary between experiments. Have the authors verified that the flow rate is consistent between experiments? Also, the VOCs are collected in integrated samples, so the duration of sampling would matter too, which may vary depending on cooking times. I looked at the paper referenced (Zhang et al, ES&TL 2021) and it seems like cooking times are ~60 min and the sampling times are ~90min, but the flow rates are not known.

Even if the air flow rate is controlled, it is difficult to compare these numbers to other experiments in the literature. I myself have gone through the literature and tried to compare different studies, but the flow rate is often not reported. I think that intensive variables, such as emission factor (ug/g of oil used) or emission flux (ug/hour) would be more useful for comparison than air concentrations.

Thank you for your comment. Unlike vehicular emissions, there was no common sense about the emission rate or emission factor of cooking emissions (Atamaleki et al., 2021). Some studies indeed utilized mass concentration (μ g m⁻³) to demonstrate cooking emissions (Huang et al., 2020). We agree that the mass concentration varies between experiments and the flow rate of cooking fumes are unknown. We convert the mass concentration into emission rates (μ g min⁻¹) for a better description of cooking emissions. Following is the revised sentence in the manuscript.

Emission rate (ER, μ g min⁻¹) was calculated by the following equation, where *c* is the blank subtracted mass concentration (μ g m⁻³) of the chemical quantified, and *Q* is the mass flow of cooking exhaust emissions (15 m³ min⁻¹).

46

$$ER = c \times Q \tag{1}$$

We did the data treatment again and the results of ER, OFP, and SOA are all presented in the mass
unit of µg min⁻¹.

49 Besides, we want to point out that though the cooking simulation experiments were conducted 50 simultaneously, we sampled Tenax TA tubes without dilution, while (Zhang et al., 2021) sampled 51 from diluted cooking fumes (dilution ratio: 8). The on- and off-line experiments were conducted 52 *separately*. Besides, the sampling time in this work is 15 ~ 30 min, not an hour.

53

In a related point, I am wondering how the quartz filter in front of the Tenax TA tubes affect the measurements, especially for the I/SVOCs. There are well known positive and negative artifacts for quartz filters, especially at high particle loadings. Some of the gaseous SVOCs can be lost to sorption 57 onto filters (or organic material on filters), and some particle phase SVOCs could evaporate off the 58 filter. What is the typical particle loading on these filters, and what is the potential for these artifacts 59 to affect the SVOC measurements. This may be especially important for SOA estimation, if SVOCs 60 contribute significantly to SOA.

61 Thank you for your comment. The emission patterns of particulate matters has been discussed in a 62 another paper (Gong, Y., Song, K., Guo, S., Lv, D., Zhang, Y., Wan, Z., Zhu, W., Wang, H., Yu, Y., Tan, R., Shen, R., Lu, S., Li, S., and Chen, Y.: Technical note: Identification and quantification of 63 64 gaseous and particulate organic compounds from cooking fumes by comprehensive two-dimensional 65 chromatography-mass spectrometry, Atmos. Chem. Phys. Discuss. [preprint], gas https://doi.org/10.5194/acp-2022-326, in review, 2022.). The total mass of particulate organics (29 66 mg m⁻³) was much larger than the total mass of gaseous organics (2.4 mg m⁻³). The particles mainly 67 contains long-chain alkanoic and alkenoic acids (linoleic acid, oleic acid, and C16-mono-acid) 68 69 covering 82.5% of the total mass. The overlap of gas- and particle-phase organics are indeed small. 70 We added the uncertainty analysis in the manuscript as follows.

71 Cooking fumes were sampled directly without dilution. After collecting particles on quartz filters, gas-phase organics were sampled by pre-conditioned Tenax TA tubes (Gerstel 6 mm 97 OD, 4.5 72 73 mm ID glass tube filled with ~ 290 mg Tenax TA) with a flow of 0.5 L min⁻¹. The 74 removal of particles on the quartz filter in front of the Tenax TA tubes affects the S/IVOC 75 measurements, causing positive and negative artifacts. Some of the gaseous SVOCs could be lost to 76 sorption onto filters, and some particle-phase SVOCs could evaporate off the filter. The emission 77 pattern of the particulate organics diverged from gas-phase organics, and a small overlap of species is 78 identified. Aromatics, aldehydes, and short-chain acids mainly occurred in the gas-phase. For 79 instance, the detection of short-chain olefinic aldehydes in the gas-phase was 40 times that of the particle-phase aldehydes. The artifacts of particulates on gas-phase aromatics and oxygenated 80 81 compounds could be less than 5%. A typical system blank chromatogram is displayed in Figure S1. A 82 daily blank sampling of the air in the kitchen ventilator was conducted before cooking and was 83 subtracted in the quantification procedure. All samples were frozen at -20°C before analyzing.

I am curious about the oil composition itself. Seems like it might be fairly straightforward to directly analyze the oil used, especially when answering the question about the differences in saturated and unsaturated fatty acid abundance. The type of oil (corn vs soybean vs other types) might not be as informative as the actual oil composition. Just a suggestion that would help add depth to the discussion, but I understand this will entail more experiments, so I will leave this up to the authors to decide whether this may be useful.

91 Thank you for your comment. We entailed a supplemental TD-GC×GC-MS experiment and added
92 the information on oil molecular compositions in the manuscript.

Quartz filters added with about 1 mL of edible oils were also thermally desorbed and analyzed
by TD-GC×GC-qMS. The total responses of blobs are normalized to 1 and the results were given by
percent response (%).

96 ...

Aromatic contributed 23.6%, 20.1%, 50.5%, and 19.8% of the total ERs of fried chicken fumes 97 98 cooked with corn, peanut, soybean, and sunflower, oils, respectively. Fried chicken fumes cooked 99 with soybean oil were especially abundant in toluene (rank 1st). In the TD-GC×GC-MS analysis of 100 soybean oil (Figure S10), unsaturated fatty acids (linoleic acid) contributed 31.5% of the total 101 percent response (50.5% aromatics), compared to 10.1% of the total response in corn oil (15.5% 102 aromatics). As a result, the aromatic concentrations and compositions of the fried chicken fumes 103 diverged according to the content of unsaturated fatty acids in the oil (Chow, 2007; Zhang et al., 104 2019).

105 ...

Although pollutants were dominated by aromatics, alkanes, and oxygenated compounds with volatility bins of B9 to B12 (VOC-IVOC range, saturated vapor concentration > $10^6 \ \mu g \ m^{-3}$) and polarity bins of P1 to P5 (low to medium polarity), significant diversities of volatility-polarity distributions were observed (Figure S9). The chemical compositions in each volatility bin were also distinct (Figure S11). IVOCs accounted for as much as 22.8% and 23.7% of the total ERs when peanut and sunflower oils were utilized for frying (Kostik et al., 2013; Ryan et al., 2008). The peanut oil was much more abundant in oleic acid (41.5%), while the proportion of linoleic acid in sunflower

- 113 is 36.6% (Figure S10). The proportion of unsaturated acids in peanut and sunflower oils is higher
- 114 than that of other oils.
- 115







I am also wondering how to interpret the main observations in the two different contexts: detailed composition and volatility/polarity distributions. The latter is a reductive approach to interpret complex organic composition, so it is not surprising that there can be larger differences in the composition (e.g. functional groups) between different experiments while the bulk volatility/polarity distribution stays relatively constant. Given the extensive analytical work performed in this study, it may be useful to dig deeper into what the composition changes can tell us. For example, is changing the oil changing the carbon number of the compounds (thereby changing the volatility distribution) whereas the cooking style only changes the functional group (and perhaps replacing one functionalgroup with another does not really impact volatility/polarity)?

Thank you for your comment. Volatility bins are commonly utilized in one-dimensional GC-MS. We want to stress that even though the volatility-polarity distribution is similar, the chemical composition could be largely different. We add figures elucidating the chemical compositions in each volatility bin in Figure S5 and Figure 12. We also revised the manuscript as follows.

133 Although the profiles of compositions diverged from dish to dish, their volatility-polarity 134 patterns remained similar. The volatility-polarity distributions of the gaseous emissions are displayed in Figure 3. VOCs (B11 and before, saturated vapor concentration $> 10^6 \ \mu g \ m^{-3}$) with low polarity 135 136 (P1 – P4) dominated the emissions of gas-phase contaminants. Chemicals in the VOC range 137 accounted for 88.7%, 95.6%, 85.2%, and 81.4% of the total emission rates of fried chicken, Kung 138 Pao chicken, pan-fried tofu, and stir-fried cabbage emissions, while S/IVOCs accounted for 11.3%, 139 4.4%, 14.8%, and 18.2%, respectively. However, considering the chemical compositions in each 140 volatility bin, the emission patterns are quite distinct (Figure S5). Oxygenated compounds were 141 widely detected before B13 (VOC-IVOC range) in emissions of fried chicken and pan-fried tofu, 142 while aromatics were extensively detected in the B8 range of Kung Pao chicken fumes. Alkanes and 143 alkenes in the B10 range dominated the emissions of stir-fried cabbage. From the discussion above, 144 the volatility distribution of cooking emissions obtained from the one-dimensional GC-MS analysis 145 faces large uncertainty in SOA estimation if the polarity is not taken into account. Meanwhile, the 146 volatility-polarity distribution should be equipped with detailed chemical parameters in each bin to 147 precisely estimate SOA.



150 Figure S5. Chemical composition-volatility distributions of four dish emissions.

Although pollutants were dominated by aromatics, alkanes, and oxygenated compounds with 152 volatility bins of B9 to B12 (VOC-IVOC range, saturated vapor concentration > $10^6 \ \mu g \ m^{-3}$) and 153 154 polarity bins of P1 to P5 (low to medium polarity), significant diversities of volatility-polarity 155 distributions were observed (Figure S9). The chemical compositions in each volatility bin were also 156 distinct (Figure S11). IVOCs accounted for as much as 22.8% and 23.7% of the total ERs when 157 peanut and sunflower oils were utilized for frying (Kostik et al., 2013; Ryan et al., 2008). The peanut 158 oil was much more abundant in oleic acid (41.5%), while the proportion of linoleic acid in sunflower 159 is 36.6% (Figure S10). The proportion of unsaturated acids in peanut and sunflower oils is higher 160 than that of other oils.



Figure S11. Chemical composition-volatility distributions of fried chicken emission cooked with fouredible oils.

166 Similar to the previous comment, the authors made a claim in the concluding section:

167 The PLS-DA and MPCA analysis indicated the importance of edible oils on cooking emissions. If 168 cooking-related pollution control strategies are made, the suggestion of deduction of oils that contain 169 more unsaturated fatty acids (such as soybean oil) could be taken into consideration.

170 It seems to me that the conclusions from the PLS-DA and MPCA analysis concern the relative 171 distributions, rather than absolute emissions. In other words, the analysis only tells you that the oil 172 determine the variation in chemical composition, but not necessarily the amount of emissions. I do 173 not disagree with the claim made in the manuscript; the evidence provided just does not support this 174 claim.

175 Thank you for your comment. We have deleted the statement in the concluding section.

There also needs to be some discussion about the limitations of GC methods to comprehensively measure all compounds. Acids can decompose during thermal desorption, if no derivatization was performed. Highly polar compounds do not elute from the GC column. This may lead to biases in estimating polarity distributions.

180 Thank you for your comment. We add some uncertainty discussions in the *Atmospheric Implications*181 parts as follows.

182 We still need to stress that although GC×GC is utilized, UCMs still occur sharing a proportion of 183 5% of the total response in this work. Acids and aldehydes tail in the second column and cause 184 quantification procedure. Meanwhile, TD-GC×GC-MS does not uncertainties in the 185 comprehensively measure all compounds. Acids can decompose during thermal desorption if no 186 derivatization was performed. Meanwhile, the decomposition of SVOC compounds could produce 187 small molecules in the VOC or IVOC range. The TD process could introduce underestimation for 188 SVOC compounds while causing overestimations of VOC and IVOC species. Highly polar 189 compounds do not elute from the GC column. This may lead to biases in estimating volatility and polarity distributions. Comparisons between GC×GC and chemical ionization mass spectrometers 190 191 (CIMS) should be further implemented to give a full glimpse of cooking organic compounds.

192

This work appears to be related to Zhang et al. ES&T 2021. How do the estimated SOA trends compare to AMS measurements? If the authors are able to reconcile SOA formation from AMS with bottom-up estimates from this work, it would allow us to assess how much we understand SOA formation in this system.

Thank you for your comment. This work is indeed related to Zhang et al. ES&T 2021 sharing the same lab, cooking material, cooking procedures, and edible oils. However, we sampled Tenax TA tubes without dilution, while (Zhang et al., 2021) sampled from diluted cooking fumes (dilution factor of 8). Besides, the on- and off-line experiments were conducted *separately*. The comparison between bottom-up and top-down methods is currently not available. However, in our recent publication (Yu et al., 2022), S/IVOCs are quantified by online VOCUS-PTR-ToF and the data were compared to AMS apportionment. VOCs could only explain 5% - 32% of the SOA, while this

- 204 percentage rises to 19% 55% when considering S/IVOC oxidation.
- 205

As mentioned earlier, I often find it difficult to understand what is being conveyed. The language in this manuscript is often confusing and awkward. There are also many instances of informal language that, in my view, is not consistent with scientific writing (e.g. "... is a tough job", "...better figure out..."). Furthermore, the number of significant figures in reported values is incongruent with the levels of uncertainty. While I will try to point out these instances of awkward language and inconsistent significant figures as much as I can in my detailed comments, there are far more than I can point out individually, and much work is needed to resolve these issues.

- 213 Thank you for your comment. We go through the text carefully and have asked a native speaker to
- 214 improve our writing.
- 215
- 216 Detailed comments:
- 217 Line 21: VOCs (not just S/IVOCs) are analyzed in this work too.
- 218 Thank you for your comment. The sentence is revised as follows.
- Volatile organic compounds (VOCs), intermediate volatility, and semi-volatile organic compounds
 (I/SVOCs) from cooking fumes were analyzed by a thermal desorption comprehensive
 two-dimensional gas chromatography coupled with quadrupole mass spectrometer
 (TD-GC×GC-qMS).
- 223
- Line 66: "clarified" is an awkward word choice.
- 225 Thank you for your comment. The sentence is revised as follows.
- 226 Although chemical compositions, fingerprints, and influencing factors of cooking emissions have
- been **investigated** in some previous studies (Alves et al., 2021; Klein et al., 2016; Peng et al., 2017;
- 228 Vicente et al., 2021), there are still questions that remain uncertain.
- 229
- Line 68: "constrain" is a verb, not a noun.
- 231 Thank you for your comment. The sentence is revised as follows.

232 The first **constraint** is that resolving complex mixtures of cooking emissions is rather tough.

233

234 Line 71: I am curious how speciating the UCM using GCxGC helped improved SOA estimation. In 235 previous work, UCM is assigned SOA yields based on total signal and prescribed volatilities. So if 236 that approach were used in this work, how different would that be from the more resolved estimates? 237 Thank you for your comment. Previous work has discussed the uncertainty of SOA estimation 238 introduced by the bins-based method. The sentence in the manuscript is revised as follows. 239 It is of vital importance to identify chemical compositions of unresolved complex mixtures (UCM) to 240 better understand their contributions to secondary organic aerosol (SOA). For instance, Huo et al 241 investigated the S/IVOC emissions from incomplete combustion utilizing GC×GC-MS. They found 242 that the previous bins-based method caused SOA underestimation with the ratio of $62.5 \pm 25.2\%$ to 243 $80.9 \pm 2.8\%$ (Huo et al., 2021). 244 245 Line 73: "ones" is an awkward word choice. Thank you for your comment. We have revised the manuscript as follows. 246 247 Particle-phase SVOC organics from cooking emissions are widely demonstrated yet few studies 248 focus on gas-phase IVOC or SVOC organics. 249 250 Line 77: I am not sure that is quite true. The canonical studies from food cooking by Schauer et al. 251 present very comprehensive profiles (Schauer et al., ES&T 1999). Thank you for your comment. We have revised the manuscript as follows. 252 253 In other words, currently, there are few comprehensive source profiles of cooking emissions covering 254 VOCs, IVOCs, and SVOCs (Schauer et al., 1999; Yu et al., 2022). 255 256 Line 112 and elsewhere: "comprehend" is not the correct word choice. Consider "understand" or 257 "study". Thank you for your comment. We have revised the manuscript as follows. 258 259 Meanwhile, four types of oil (i.e., soybean, corn, sunflower, and peanut oil) were used for frying 260 chicken to **illustrate** the influence of oil.

261

Line 115: it is slightly confusing to say that the emissions are mixed with ambient air (which is essentially dilution) and then say measured without dilution.

Thank you for your comment. We have deleted the statement about mixing and revised the manuscript as follows.

- 266 Cooking fumes were sampled directly without dilution.
- 267

Line 117: what are the breakthrough volumes of the most volatile compounds on the Tenax tubes?

269 0.5L/min for 90 minutes is about 45L. Are there concerns about compound breakthrough?

270 Thank you for your comment.

We did a supplementary experiment to examine the breakthrough effect by introducing pure nitrogen gas to the desorption tube with pre-added standard chemicals (Figure SS1). No significant breakthrough was observed within 24 h (<3%). The sampling time in this work is $15 \sim 30 \text{ min} (0.5 \text{ L} \text{ min}^{-1})$ which is much less than 24h. We revised the manuscript as follows.

A Tenax TA breakthrough experiment was conducted by introducing pure nitrogen gas (N₂) with a flow of 0.5 L min⁻¹ to the desorption tube with pre-added standard chemicals (Figure S2). No significant breakthrough was observed within 24 h (<3% of TIC). The sampling time in this work is $15 \sim 30 \text{ min} (0.5 \text{ L min}^{-1})$ which is much less than 24h.



- Figure S2. The chromatograms of standard chemicals after 6h (brown), 24h (blue), 48h (red), and 72h (blue) of flowing by pure nitrogen gas. The flow of nitrogen gas is set to be the same as the sampling flow (0.5 Lmin^{-1}). No significant breakthrough was observed within 24 h (<3%).
- 283
- Line 130: how good is the assumption that the 1st dimension retention time is representative of volatility? Did the authors verify against calculated vapor pressures?
- 286 Thank you for your comment. Pure liquid vapor pressure (p_L, P_a) has been estimated by pixel-based
- approaches in our previous publication (Song et al., 2022), which validates our statement.
- 288
- Line 132: what does "qualified" mean?

290 Thank you for your comment. We have deleted the word as follows.

291 326 chemicals were quantified (Table S3) while 201 contaminants were detected (Table S4) in

292 cooking fumes covering a wide range of VOCs, IVOCs, and SVOCs, including 25 aromatics, 19

n-alkanes, 100 oxygenated compounds (containing 7 acids, 10 alcohols, 29 aldehydes, 24 esters, 5

- ketones, and others), 3 PAHs, and 54 other chemicals.
- 295
- Line 132-133: "kinds" is an awkward word choice.
- 297 Thank you for your comment. We have revised the manuscript as follows.
- 298 326 chemicals were quantified (Table S3) while 201 contaminants were detected (Table S4) in
- 299 cooking fumes covering a wide range of VOCs, IVOCs, and SVOCs, including 25 aromatics, 19
- 300 *n*-alkanes, 100 oxygenated compounds (containing 7 acids, 10 alcohols, 29 aldehydes, 24 esters, 5
- 301 ketones, and others), 3 PAHs, and 54 other chemicals.
- 302
- Line 167: the word "form" is repeated. Also, I think the authors mean "format"?
- 304 Thank you for your comment. NetCDF is the abbreviation of Network Common Data Form. We have
- 305 revised the manuscript as follows.
- 306 Chromatograms were imported from the network common data form (netCDF).

- 308 Line 190-193: how do these numbers compare to other works?
- 309 Thank you for your comment. We add a comparison as follows.

310 The compositions of the gaseous emissions are exhibited in Figure S4. Aromatics contributed 311 59.1%, 23.6%, 8.1%, and 11.8% of the total mass concentration of Kung Pao chicken, fried chicken, 312 pan-fried tofu, and stir-fried cabbage, while oxygenated compounds accounted for 17.1%, 53.7%, 313 76.9%, and 25.0% of the total concentration, respectively. The compositions of organic in this study 314 diverged from proton transfer reaction mass spectrometer (PTR-MS) measurements (Klein et al., 315 2016; Liu et al., 2018), in which aldehydes dominated the emission profiles (~ 60%). The proportion 316 of aromatics was also different from online Vocus-PTR-ToF measurements in a recent study (Yu et 317 al., 2022). However, the contribution of aromatics was close to a recent study conducted at Chinese 318 restaurants using GC-MS analysis (Huang et al., 2020). The different instruments resulting in 319 different VOC detection ranges could be the explanation for the different patterns. GC×GC-MS is 320 powerful in resolving complex mixtures with carbon numbers of more than 6. The structural 321 chromatograms and detailed mass spectrum information provide a convincing result in chemical 322 identification (An et al., 2021). In contrast, PTR-MS could detect much more short-chain alkenes and 323 aldehydes with carbon numbers less than 4. However, the isomers of PTR-MS could not be 324 distinguished. Alkanes and some long-chain compounds could not be detected by PTR-MS. For 325 instance, the maximum carbon number of pollutants in Yu et al is 16 ($C_{16}H_{26}$) (Yu et al., 2022) while 326 the maximum carbon number of pollutants detected in this work is 30 (C₃₀H₆₂). C₂H₆O, C₄H₈. 327 C₄H₈O2, and C₅H₈ were the top species measured by Vocus-PTR-ToF (Yu et al., 2022), which is out 328 of range of our measurement.

329

Line 209: It is more common in this field to use saturation vapor pressure or saturation concentrations to denote volatility, and O/C for polarity. What are the equivalent c* and O/C for these bins?

333 Thank you for your comment. We add the saturation concentrations in brackets as follows.

334 The volatility-polarity distributions of the gaseous emissions are displayed in Figure 3. VOCs (B11

and before, saturated vapor concentration > $10^6 \ \mu g \ m^{-3}$) with low polarity (P1 – P4) dominated the

336 emissions of gas-phase contaminants.

- 337
- Line 268-273: this paragraph is confusing. It may be helpful to have a sentence suggesting that this
- 339 paragraph will be discussing the oil effect, rather than opening with "As for OFP estimation..."
- 340 Thank you for your comment. We revised the manuscript as follows.
- 341 Chicken fried in soybean oil produced the highest OFP ($10134 \pm 5958 \ \mu g \ min^{-1}$) while chicken fried
- in corn oil resulted in the most SOA estimation $(426 \pm 270 \ \mu g \ min^{-1})$.
- 343
- 344 Line 277: typo in "short-chain"
- 345 Thank you for your comment. We revised the manuscript as follows.
- 346 Despite the importance of aldehydes revealed in previous studies (Klein et al., 2016; Liu et al., 2018),
- 347 our results demonstrated that alkanes, pinenes, and short-chain acids are also key precursors in
- 348 cooking SOA production (Huang et al., 2020).
- 349

Line 279: what are "key reactions"? Is this referring to in-oil reactions? I am not sure if this study is really elucidating these reactions. Almost all cooking emission studies do not measure oil composition directly, and are only inferring these reactions based on food science literature. It is unclear if these measurements help elucidate these reactions.

- Thank you for your comment. We revised the subtitle as follows.
- 355 3.3 Elucidating the influencing factor and **inferring** in-oil reactions of cooking emissions
- 356
- 357 Line 289: typo in "variance"
- 358 Thank you for your comment. We revised the manuscript as follows.
- 359 The **variance** of cooking fumes could be largely explained by the different oil utilized.
- 360

Line 304-306: this is an interesting point. Did the emissions of aromatics increase with degree of unsaturation in oil?

363 Thank you for your comment. The emissions of aromatics decrease with the decreasing degree of

unsaturation in oil. We revised the subtitle as follows.

In more detail, the oxidation of unsaturated fatty acids (such as linoleic acid) in oil leads to the production of alkadienals (such as (E, E)-2,4-decadienal) which form aromatics (butylbenzene) by losing H₂O (Atamaleki et al., 2021; Zhang et al., 2019). This is consistent with the analysis of edible oils in this work. The emission pattern is in line with previous studies (Atamaleki et al., 2021). Corn oil contained a less amount of unsaturated fatty acids (Figure S10), and the emission of aromatics cooked with corn oil was the lowest among the 4 types of oils used. The emission pattern is in line with previous studies (Atamaleki et al., 2021).

372

373 Section 4: the conclusion section is more a recap of the results and discussion, and very thin on 374 implications and limitations. I suggest a broader discussion of context, and posing future research 375 questions.

Thank you for your comment. We revised section 4 as follows.

377 In this work, gaseous VOCs, IVOCs, and SVOCs from cooking fumes are quantified in detail. 378 The influence of cooking style and oil is taken into account in this work. S/IVOC species are key 379 components as they contributed 10.2% - 32.0% of the total SOA estimation. Previous works might 380 underestimate the importance of cooking fumes to SOA formation because only a series of IVOC 381 homologs were quantified (Liu et al., 2018). For instance, aldehydes only accounted for 0.7% -10.1% 382 of the total SOA estimation. If only aldehydes are taken into consideration, SOA will be 383 underestimated 9.9 ~ 139 times. We still need to stress that although GC×GC is utilized, UCMs still 384 occur sharing a proportion of 5% of the total response in this work. Acids and aldehydes tail in the 385 second column and cause uncertainties in the quantification procedure. Meanwhile, TD-GC×GC-MS 386 does not comprehensively measure all compounds. Acids can decompose during thermal desorption 387 if no derivatization was performed. Meanwhile, the decomposition of SVOC compounds could 388 produce small molecules in the VOC or IVOC range. The TD process could introduce 389 underestimation for SVOC compounds while causing overestimations of VOC and IVOC species. 390 Highly polar compounds do not elute from the GC column. This may lead to biases in estimating volatility and polarity distributions. Comparisons between GC×GC and chemical ionization mass 391

392 spectrometers (CIMS) should be further implemented to give a full glimpse of cooking organic393 compounds.

394 We also first proposed a novel two-dimensional panel elucidating the physiochemical properties 395 of contaminants from the perspectives of their volatilities and polarities. This novel scheme is 396 appropriate to demonstrate the complicated evolution of contaminants clearly and provide new 397 insight into the previously 1D-bins method. The volatility-polarity panel inherited the spirit of the 398 two-dimensional volatility-based set (2D-VBS) (Donahue et al., 2011, 2012) and would be further 399 implemented in the analysis of complex ambient or source samples along with the powerful 400 separating capacity of GC×GC. We would like to emphasize the importance of combining the 401 volatility-polarity distribution with detailed chemical information for a precise estimation of SOA.

We also provide powerful tools in speciating the main driving factor and inferring chemical reactions in rather complicated systems. The pixel-based PLS-DA and MPCA analysis greatly enhance our learning of complex chromatograms and provide us with new insight into the dimension reduction processes. The analyzing scheme could benefit those analysts with less experience in GC×GC data processing.

407 Our results demonstrated that both cooking styles (dish) and oils influence the cooking 408 emissions. Kung Pao chicken emitted more pollutants than other dishes due to its rather intense 409 cooking method. Cooking materials could also influence the compositions of fumes as well. 410 Aromatics and oxygenated compounds were extensively detected among meat-related cooking fumes, 411 while a vegetable-related pattern was observed in the emissions of stir-fried cabbage. As much as 412 22.2% and 29.5% of the total organics of stir-fried cabbage emission were alkanes and alkenes 413 (especially pinenes). On the other hand, oils greatly influence the composition and volatility-polarity 414 distribution of pollutants. Chicken fried with corn oil emitted the most abundant contaminants. 415 However, the ozone formation from soybean-oil fried chicken fumes was much higher. Considering 416 the high consumption proportion of soybean oil (~ 44% in volume of oil usage) in China (Jamet and 417 Chaumet, 2016), the influence of using soybean cooking oil on ozone formation might be 418 underestimated. The MPCA results also indicate that the heating and cooking procedure greatly 419 enhances the autooxidation of oil. MPCA results emphasize the importance of the unsaturated fatty

420	acid-alkadienal-volatile product mechanism. N	More studies	need to	be carried o	n to elucidate	the key
421	chemical reactions between the food and oil.					

- 422
- 423 Line 322-323: the authors can substantiate this claim with much more quantitative information. How
- 424 much of the estimated SOA is from aldehydes versus other compounds based on the calculated SOA
- 425 formation potential (equation 2)?
- 426 Thank you for your comment. We revised the manuscript as follows.
- 427 The influence of cooking style and oil is taken into consideration in this work. S/IVOC species are
 428 key components as they contributed 10.2% 32.0% of the total SOA estimation.
- 429 Aldehydes only accounted for 0.6% -10.1% of the total SOA estimation. We revised the manuscript430 as follows.
- 431 Previous works might underestimate the importance of cooking fumes to SOA formation because
- 432 only a series of IVOC homologs were quantified (Liu et al., 2018). For instance, aldehydes only
- 433 accounted for 0.6% -10.1% of the total SOA estimation. If only aldehydes are taken into
- 434 consideration, SOA will be underestimated 9.9 ~139 times.
- 435
- 436 Supplemental Information:
- 437 Table S1: how were oil temperatures measured or estimated?
- 438 Thank you for your comment. The oil temperature was measured by a thermometer placed in the oil.
- 439 The thermometer was removed from the oil before placing the cooking materials. We revised Table
- 440 S1 as follows.
- 441 **Table S1.** Details of cooking procedures.

Domestic	Material	Oil	
cooking		temperature #	
Fried chicken	170 g chicken, 500 mL oil (corn, peanut, soybean, or sunflower	145 ~	
	oil), a few condiments	150 °C	
Kung Pao	150 g chicken, 50 g peanut, 40 mL corn oil, a few condiments	Not stable	
chicken			

	Pan-fried tofu	500 g tofu, 200 mL corn oil, a few condiments	100 ~			
			110 °C			
	Stir-fried	300 g chicken, 40 mL corn oil, a few condiments	95 ~ 105 °C			
	cabbage					
442	[#] The oil temperature	e was measured by a thermometer placed in the oil. The	thermometer was			
443	removed from the oil	before placing the cooking materials. The temperatures liste	d in Table S1 were			
444	initial cooking temperatures and were maintained the same for each dish.					
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