

Atmos. Chem. Phys. Discuss., referee comment RC1
<https://doi.org/10.5194/acp-2022-320-RC1>, 2022
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Comment on acp-2022-320

Anonymous Referee #1

Referee comment on "Impact of cooking style and oil on semi-volatile and intermediate volatility organic compound emissions from Chinese domestic cooking" by Kai Song et al., Atmos. Chem. Phys. Discuss., <https://doi.org/10.5194/acp-2022-320-RC1>, 2022

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

Major comments:

All of the emissions are reported in air concentrations ($\mu\text{g per m}^3$ 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 ~ 90 min, 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.

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 onto filters (or organic material on filters), and some particle phase SVOCs could evaporate off the filter. What is the typical particle loading on these filters, and what is the potential for these artifacts to affect the SVOC measurements. This may be especially important for SOA estimation, if SVOCs contribute significantly to SOA.

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.

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 functional group with another does not really impact volatility/polarity)?

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

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

It seems to me that the conclusions from the PLS-DA and MPCA analysis concern the

relative distributions, rather than absolute emissions. In other words, the analysis only tells you that the oil determine the variation in chemical composition, but not necessarily the amount of emissions. I do not disagree with the claim made in the manuscript; the evidence provided just does not support this claim.

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.

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.

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.

Detailed comments:

Line 21: VOCs (not just S/IVOCs) are analyzed in this work too.

Line 66: "clarified" is an awkward word choice.

Line 68: "constrain" is a verb, not a noun.

Line 71: I am curious how speciating the UCM using GCxGC helped improved SOA estimation. In previous work, UCM is assigned SOA yields based on total signal and prescribed volatilities. So if that approach were used in this work, how different would that be from the more resolved estimates?

Line 73: "ones" is an awkward word choice.

Line 77: I am not sure that is quite true. The canonical studies from food cooking by Schauer et al. present very comprehensive profiles (Schauer et al., ES&T 1999).

Line 112 and elsewhere: "comprehend" is not the correct word choice. Consider "understand" or "study".

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.

Line 117: what are the breakthrough volumes of the most volatile compounds on the Tenax tubes? 0.5L/min for 90 minutes is about 45L. Are there concerns about compound breakthrough?

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?

Line 132: what does "qualified" mean?

Line 132-133: "kinds" is an awkward word choice.

Line 167: the word "form" is repeated. Also, I think the authors mean "format"?

Line 190-193: how do these numbers compare to other works?

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?

Line 268-273: this paragraph is confusing. It may be helpful to have a sentence suggesting that this paragraph will be discussing the oil effect, rather than opening with

"As for OFP estimation..."

Line 277: typo in "short-chain"

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.

Line 289: typo in "variance"

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

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

Line 322-323: the authors can substantiate this claim with much more quantitative information. How much of the estimated SOA is from aldehydes versus other compounds based on the calculated SOA formation potential (equation 2)?

Supplemental Information:

Table S1: how were oil temperatures measured or estimated?

References:

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