

Comment on acp-2022-326

Anonymous Referee #1

Referee comment on "Technical note: Identification and quantification of gaseous and particulate organic compounds from cooking fumes by comprehensive two-dimensional gas chromatography-mass spectrometry" by Yuanzheng Gong et al., Atmos. Chem. Phys. Discuss., <https://doi.org/10.5194/acp-2022-326-RC1>, 2022

In this work, the authors measured organic compounds emitted from cooking fumes using thermal desorption multidimensional gas chromatography mass spectrometry (TD-GCxGC-MS). This technique has gained popularity for analysis of complex mixtures, since it increases the separation capacity and provide more compound resolution. The thermal desorption technique also allows for parallel analysis of particle and gas phase samples. The authors report that 170-350 compounds were identified, which represented over 90% of the elutable signal.

According to ACP policy, a technical report would "report new developments, significant advances, or novel aspects of experimental and theoretical methods and techniques that are relevant for scientific investigations within the scope of the journal." In my view, this work does not meet any of these criteria, and the work does not advance scientific understanding of food cooking fume in any significant way. Furthermore, the manuscript misrepresents the usefulness of the TD-GCxGC-MS and does not address any of its limitations, some of which are evident from the data presented. The analysis is filled with errors, including incorrect grouping of compounds. The manuscript is poorly written and difficult to understand. I recommend rejection of this manuscript.

Major comments:

My biggest issue with this work is that almost all of the compounds have been reported in previous literature on food cooking emissions. Many of the observations from this technique have long been established since the 1990s starting with work by Rogge et al. As I browsed through the list of compounds in the Supplementary Material, most of the

compounds have been reported by previous studies. While there are new compounds reported here, I am not sure if this is valuable information: it is not clear how they are related to the cooking process (e.g. sulfur dioxide? Dotriacontyl trifluoroacetate?); many are very minor in concentrations. A simple list of compound generated by the analysis software without useful interpretation is not useful to any reader and adds no value to the literature.

I also have some technical concerns about the methods. The flow is split into two, one to collect gas phase compounds on Tenax, and the other to collect particles on quartz filters. Quartz filters are known to experience positive artifacts, i.e. gas phase SVOCs can adsorb onto quartz filters. Would there be double counting of SVOCs for this reason? The authors did not attempt to address these artifacts.

The statement that 95%/90% of the total signal was speciated is somewhat misleading. The total signal represents what elutes from the GC columns but not total organic mass. One of the main issue with GC techniques is that much of the material does not elute from the column, as the polar compounds either decompose or partition irreversibly to the stationary phase of the column. The most appropriate way to make this comparison is to compare against total PM mass (for the quartz filter samples). One way to get total OC mass on the filters would be OC/EC analysis. That would be a much fairer comparison.

The manuscript is written very poorly and is hard to understand. It is riddled with spelling mistakes, punctuation errors and awkward language.

Table S1 in Supplementary Material (which I think is the crux of this work) is poorly presented. The grouping of compounds is inconsistent and in many cases simply wrong. Here are just some examples:

- d-mannose (listed under "Aldehydes/Saturated Aldehydes") is not an aldehyde.
- "1-Hexanol, 2-ethyl-" is listed as "Others", but should be in "Alcohols".
- "2-Pentadecanone" listed under "Esters and Amides" but should be in "ketones"

Other comments:

Introduction: Lines 38-40 and lines 57-58 both describe COA fraction, but the numbers cited are all over the place. I understand that they likely have different contexts (e.g. using different source apportionment methods), but as it stands it does not set the proper

stage for this work.

Lines 72-74: this is a misrepresentation of the state of the science. Rogge et al. and Schauer et al. showed a much greater variety of organic compounds that can be identified and quantified.

Lines 79-89: I would think that previous studies using GCxGC-MS for analysis of organic compounds in the atmosphere should be reviewed here, which will demonstrate their capabilities (and limitations).

Section 2 Methods: Some details regarding optimizing GCxGC would be useful. E.g. what is the data acquisition rate of the mass spectrometer? Usually you would need fast data acquisition to properly define a peak shape in the 2nd dimension (at least 10 per peak). Also, what is the number of cuts across each peak in the 1st dimension? Usually you would want to have at least 3 cuts across each peak. Given the slow temperature ramp (5C/min) I do not suspect this to be a problem, but it would be good to show.

Line 120: CIS has already been defined

Line 161: the difference in RI between the compounds is bigger than the difference in RI between experimental and literature, which makes this identification not convincing. For aromatic compounds, Lee's RI may be more appropriate.

Line 203: this sentence is simply not true, and is stated without evidence. There is UCM in Table S1, only 90-95% was identified, and no comparison with total mass.

Figure 1: there is significant tailing for acids. This is not surprising given their polarity and the nonpolar compounds used as the 1st dimension. However, this limitation is not discussed at all.

Line 258 (and other instances): the level of certainty is incongruent with the number of significant figures.

Lines 264-267: to be a good tracer, a compound has to be emitted uniquely from a source. D-limonene has many other atmospheric sources (biogenic emissions, indoor cleaners etc.) and would not be a good tracer.

Line 274: I think poor quantification of acids by GC or GCxGC is more likely. Acids decompose to CO₂ at GC temperatures. That is why GC analysis of organic acids often require derivatization. Acids have a higher proton affinity than water and is therefore amenable to PTR-MS analysis.

Line 285-286: again, this is simply not true! See Schauer et al and so many other papers for identification of N-containing compounds.

Line 326-327: this is to be expected since no derivatization was performed here. Previous work often use silylation to convert acidic OH groups to non-acid O-Si(CH₃)₃ groups.

Line 343-344 None of these numbers can be trusted, since the grouping of compounds was not done properly (see comment on Table S1).