

Atmos. Chem. Phys. Discuss., referee comment RC2
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Comment on acp-2021-20

Anonymous Referee #2

Referee comment on "Formation and evolution of secondary organic aerosols derived from urban-lifestyle sources: vehicle exhaust and cooking emissions" by Zirui Zhang et al., Atmos. Chem. Phys. Discuss., <https://doi.org/10.5194/acp-2021-20-RC2>, 2021

Zhang et al. measured SOA formation from emissions from a GDI engine and four types of food cooking emissions that were exposed to OH radicals in a Go:PAM. Equivalent atmospheric aging timescales of up to 5 days were studied. The SOA/POA ratio was approximately 100 for the oxidized vehicle exhaust and 2 for the oxidized cooking exhaust. Higher SOA oxidation states were observed for the oxidized vehicle exhaust. AMS spectra of the oxidized emissions were examined and compared to ambient LO-OOA, MO-OOA, and COA factors resolved using PMF. The studies are well motivated. In its current form, there are too many important experimental details that are missing for me to support publication in ACP.

Comments

- Critical details about the sampling inlets between the source emissions and the OFR (e.g. tubing length, diameter, material, residence time) that could influence the penetration efficiency and/or delays in transmission to the OFR (e.g. Pagonis et al, 2017) are missing. If these conditions vary between vehicular exhaust and cooking exhaust measurements, that is one of several potentially important variables that could complicate direct comparison of results between the two studies.
- It is not clear to me why different OFR conditions were used in the vehicle exhaust and food cooking experiments. For example, the residence time was 110 s and the RH was 44-49% in the GDI exhaust studies, compared to 55 seconds' residence time and RH = 18-23% in the food cooking studies. This makes it more challenging to directly compare results from the different studies – for example, although there may be overlap in OH exposure between the different experiments, timescales for gas-to-particle condensation and any humidity-dependent heterogenous chemistry are different.
- More detailed information about the gas-phase measurements are necessary to interpret the results. For example, it is not clear to me whether more SOA is formed from aging of vehicle emissions because the VOC concentrations are higher, the SOA yields of those VOCs are larger, or both. Please add table(s) showing the list of VOCs

that were measured from each source, their emission factors relative to CO₂, and their OH rate coefficients that were used to calculate the total external OH reactivity.

- **L96:** A field study at IAP is mentioned here, but it is not clear until much later (L254) that results from this study are (I think) already published in the Li et al. (2020a) paper that is referenced much later in the manuscript.
- **L112:** Given the presumably large emission factors of unsaturated fatty acids in the cooking emissions, and their corresponding fast reaction rate coefficients with ozone, it would have been useful to conduct control measurements to measure the ozonolysis products of the cooking emissions. Why were those experiments not performed here?
- **L136:** The information in Tables 1-2 indicates that the sample line temperature was 20-25°C, presumably at or close to room temperature. How did the authors determine that this temperature was sufficient to “prevent freshly warmed gas from condensing on the pipe wall”?
- **L137:** What are the particle backgrounds when the lamps are turned on with ozone and humidified air flowing through the Go:PAM? Simply flowing dry purified air and ozone through a dark OFR is likely insufficient to clean out the OFR between experiments that employ OH as the oxidant. In this case, the background concentrations are probably significantly underestimated because as soon as the lamps are turned on, there is the potential to generate SOA from the OH oxidation of background contaminants that are not reactive towards O₃.
- **L159:** In addition to the OFR conditions that were summarized in Tables 3-4, the actinic flux at 254 nm (or, alternatively, the ratio of O₃ measured before and after photolysis at 254 nm) is also a required input to the OFR254 OH exposure estimator. Please add this information to Tables 3-4 or describe in the text.
- **L161-L165:** This discussion is confusing and in places incorrect. I don't understand why acetylacetone is mentioned at all if it is not present in the emissions, whereas other specific VOCs that were measured are not discussed here or anywhere else in the paper. Also, I suspect ozone is likely an important oxidant for some species, especially unsaturated fatty acids that are presumably important components of the cooking emissions as discussed in the paper. For example, for oleic acid, at OH and O₃ exposures of 2.7E11 and 5E15 molecules cm⁻³ s (OFR conditions in Line 5 of Table 4), assuming effective OH and O₃ rate coefficients of 3.5E-11 and 2.1E-15 cm³ molecule⁻¹ s⁻¹ (Renbaum et al, 2012), the estimated fractional oxidative loss of oleic acid to O₃ is ~0.55. Thus, O₃ may actually be the major oxidant for several important compounds emitted from the cooking sources.
- **L203:** The O/C ratio is insufficient by itself to associate the mass spectra of the SOA with ambient PMF factors.
- **L207 and L227-L228:** These statements are too speculative, and references are made to other source characterization studies that are not directly relevant to the sources that were characterized here. Molecular speciated measurements of VOCs were performed with GC-MS as described in L168-L172 that are not discussed in the text. Those measurements should either support (or not) this interpretation of the results. And the fatty acids that are mentioned have high effective OH rate coefficients (e.g. Renbaum et al., 2012), so it is not obvious to me how the statement that “cooking produces more hardly oxidized acids” is justified.
- **L225:** Is “souring” a typo? If not, I am not certain what this statement means.
- Figures 2-3, 7. I find it confusing/distracting to have two different photochemical age and/or two SOA:POA axis scales on the same figure.
- Figure 6: I think that the numbered symbols represent the integer m/z values of the average vehicular and cooking exhaust AMS spectra, but this should be made clearer in the legend or the caption.

References

L. Renbaum-Wolff and G. D. Smith, "Virtual Injector" Flow Tube Method for Measuring Relative Rates Kinetics of Gas-Phase and Aerosol Species, *J. Phys. Chem. A*, 116, 6664–6674, [dx.doi.org/10.1021/jp303221w](https://doi.org/10.1021/jp303221w), 2012.

Pagonis, D., Krechmer, J. E., de Gouw, J., Jimenez, J. L., and Ziemann, P. J.: Effects of gas-wall partitioning in Teflon tubing and instrumentation on time-resolved measurements of gas-phase organic compounds, *Atmos. Meas. Tech.*, 10, 4687–4696, <https://doi.org/10.5194/amt-10-4687-2017>, 2017.