Review of

**"Characteristics and degradation of organic aerosols from cooking sources based on hourly observation of organic molecular markers in urban environment"**

By Li et al.

<u>General comments</u>

This study aims to characterise the cooking organic aerosol markers evolution in the atmosphere. The study employs online thermal desorption aerosol gas chromatography mass spectrometry (TAG) to analyse the organic component of PM2.5 from urban Changzhou, China. The study identified and attributed saturated fatty acids, unsaturated fatty acids, and oxidative decomposition products of unsaturated fatty acids as the molecular marker of cooking sources. Additionally, the decomposition of these markers was estimated using an established ageing parameterisation model, and the contribution of the cooking source to PM2.5 was estimated using a positive matrix factorisation model.

The online molecular analysis employed by this study gives a great deal of data that, unfortunately not being optimally used in the analysis and discussion. The study adopted and modified analysis methods by Wang et al. (2020) and Wang and Yu (2021). However, the modification is not entirely justified, raising more questions about the results and discussion. Finally, the association of fatty acids as cooking source molecular markers need more evidence/analysis that considers the other sources, such as biomass burning and marine aerosol.

The manuscript is well written, with some typos that can be improved after thorough checking. The topic presented in this study fits within the scope of the *Atmospheric Chemistry and Physics* journal. My main concerns are the insufficient evidence to support the cooking organic aerosol molecular marker characterisation and their atmospheric decomposition. In summary, I recommend the Editor reconsider the manuscript after major revisions.

<u>Specific comments</u>

1. Although following previous studies, The Methods section needs explanation and clarifications, which are as listed below.
    a. The PM2.5 sampling method (Lines 94-96: flow rate, residence time, type of collection matrix, etc)
    b. The post-sampling analysis (Lines 100-102: brief procedures). For example, an hourly sampling will result in 24 samples at maximum. An hour of sampling plus 1.5-hour post processing before starting a new sampling sequence will result in less than 12 samples per day.
2. The identification of cooking aerosol markers needs improvement. Attributing all Fatty Acids as cooking aerosol markers is misleading because they are also emitted by other sources, i.e., biomass burning and traffic. The study used a similar measurement method (TAG) to Wang et al. (2020) and is referred to it in the Methods section. That means this study measured other organic molecules, such as sugars (levoglucosan) and PAHs, similar to Wang et al. (2020) study. Moreover, the Supplement Section S2 Figure S7a shows source

profiles composed of small and long-chain acids, PAHs, and sugars, suggesting that these data are available for discussion. These other organic groups would improve the identification of cooking source contribution to the total fatty acids emissions.

a. Lines 71-74: They are not only markers for cooking/culinary emissions. They can also be emitted by biomass burning, vehicles, and plants (Rogge et al., 1991, 1993, 1998; Simoneit, 2002). Since there are a couple of potential emission sources, discuss how previous studies differentiate them (Ho et al., 2015, Wang et al., 2020). Ref: Simoneit (2002, https://doi.org/10.1016/S0883-2927(01)00061-0), Rogge et al. (1998, https://doi.org/10.1021/es960930b), Rogge et al. (1991, https://doi.org/10.1021/es00018a015), Rogge et al. (1993, https://doi.org/10.1021/es00041a007), Ho et al. (2015, doi:10.5194/acp-15-3111-2015)
b. What are EP1, EP2, etc., in Figure 2. A summary table of EP1, EP2, etc., parameters can be added to the Supplement. Additionally, plotting fatty acids in log scale make the time trends incomparable to the other parameters plotted in linear scale (Figure 2). What are the reasons behind separating EP2 and EP3 (Figures 2 and 4)? Instead of grouping and averaging the measurement into day-measurement (D1, D2, D3, etc.), time series of TFAs/OC for EP1, EP2, etc. would show the actual trend and give better insight into the transformation of the fatty acids (Lines 265-269).
c. Section 3.1: How TFAs and FAs are differentiated here? TFAs (Line 168) are similar to FAs (Line 176). Be consistent with using fatty acids or FAs, and FAs or TFAs. Additionally, Considering the other sources of fatty acids, the missing of lunchtime raises a question (Lines 180-185). Could the fatty acids come from biomass burning (residential heating) at night and vehicle emissions in the morning? Wang et al. (2020) observed a small increase in the daytime. Additionally, back trajectory analysis suggests CL#2 and CL#4 airmasses came from the sea (Line 223, Figure 5), suggesting a marine aerosol potential contribution to fatty acids.
d. Based on Figures 4 and 3, the cooking source seems to contribute at night only. In the morning, the fatty acids could be contributed from non-cooking or long-range transport sources, of which the latter can be a combination of sources. Additionally, Oleic/Palmitic in Figure 4 is peaking at noontime, which could be associated with fresh cooking emissions at lunchtime. Instead of individual species analysis, cluster analysis of the ratios of the fatty acids could give further insight into their atmospheric evolution.

3. This study used parameterisation by Wang and Yu (2021) to estimate the fatty acids atmospheric ageing. Wang and Yu (2021) developed the relative rate constant for the ambient fatty acids based on C18:0 stearic acid as the reference molecule for normalisation.
   a. Any reason for choosing Palmitic Acid instead of Stearic Acid (Lines 132-133)? Wang 2020 plotted Oleic/Stearic or Linoleic/Stearic vs Palmitic/Stearic to

investigate the ageing of cooking markers. Is there a particular reason for plotting Oleic/Palmitic or Linoleic/Palmitic versus Palmitic/Stearic (Lines 194-202, Figure 4)? Using different denominators would result in different ageing interpretations. However, it could be good to test the effect of Stearic Acid and Palmitic Acid as the ageing reference to the analysis.

   b. Lines 292-293, Figure 8: The plot does not show negative linear correlation as inferred in the text and figure. The curves could be potentially negative exponential for Azelaic or Oxononanoic/Palmitic vs Oleic/Palmitic, but not linear. Moreover, there is no R or $R^2$ value for the regression plots. The R or $R^2$ value is important to assess the association between the two ratios/parameters. Another method is calculating the p-value to show the statistical significance of the two ratios.
   c. What is the reason for plotting Figure 9 in a 2-power scale instead of a linear scale? If these axes are correlation values for X9/P vs O/P, Figure 9 should plot $R^2$ values of each ratio.

4. This study used PMF analysis to estimate cooking source contribution to PM2.5. However, this study has not adequately discussed the PMF analysis leading to the conclusion of cooking source contribution. It is mentioned that this study would only briefly identify each source factor. However, a discussion of the PMF analysis is still needed to support the conclusion on cooking aerosol contribution to PM2.5. The PMF analysis can go to the Supplement as done in this manuscript, but it needs more information. The authors should provide, at a minimum, the correlation coefficient of factor solution profiles and time series and the observed and predicted cooking aerosol markers. Further information, such as the Base Model Displacement Error Method and Bootstrapping analysis, is also important to explore the rotational ambiguity and assess the uncertainty that arises from random errors in the dataset, respectively (e.g., Almeida et al. (2020, https://doi.org/10.1016/j.envpol.2020.115199). Lastly, considering the cooking contribution is smaller than biomass burning, and vehicle exhaust (Figure 12), fatty acids discussed here might not be mainly emitted by cooking activities.

<u>Technical comments</u>

a) Lines 227-228: The component of ODPs haven't been explained in the text.
b) Figure 5: Add a legend to explain the coloured lines for the Cluster.
c) Lines 260-262: Add reference studies or ratios for TFAs/OC from other sources.
d) Figure 8a: What does the y-axis Contribution mean?
e) Line 311: Correlation as in correlation coefficient values or relationship between Y and X axes?
f) Lines 391-302, Figure S7: There is no time series or diurnal plot of the PMF factor solution in the Supplement.