

Review:

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Kaltsonoudis et al. presents a series of smog chamber experiments to study the aging of primary emissions from meat charbroiling. They found that the initial and aged AMS spectra of meat charbroiling differed considerably. The derived fresh and aged cooking factors in laboratory were compared with ambient COA factors during Fat Thursday in Patras, Greece.

The experiments are novel and performed with an extensive suite of instrumentation to systematically study the chemical aging of emissions from a very important anthropogenic source, namely meat charbroiling. The paper potentially has significant implications to the formation of SOA and control of PM in many urban environments. While the datasets are interesting, the manuscript can be improved by providing more detailed interpretation of the data. The paper can be recommended for publications after the following questions are addressed.

General comments:

1. The experiments were not conducted at atmospheric relevant conditions, which could bias the conclusions and implications. As shown in Table 1, the mass concentration of PM<sub>1</sub> ranged from 130  $\mu\text{g m}^{-3}$  to 540  $\mu\text{g m}^{-3}$ , much higher than typical concentrations of PM<sub>1</sub> in ambient air. The chemistry occurred at higher mass loadings of OA may be different from that at lower mass loadings. Did the authors conduct experiments at lower mass loadings of OA? If not, some discussions to relate the current findings to atmospheric implications at more realistic PM concentrations would be needed.

2. In Line 1-3, Page 6, the authors mentioned that “For the same sampling time the AMS mass concentration (CE=1) was 600  $\mu\text{g m}^{-3}$ , the SMPS (assuming density 1  $\text{g cm}^{-3}$ ) was 100  $\mu\text{g m}^{-3}$ , and the filter-based concentration was 500  $\mu\text{g m}^{-3}$ .” Was a CE value of 1 applied to the entire study of POA and SOA in laboratory and ambient measurements? As CE values are dependent on chemical composition of PM<sub>1</sub> (Middlebrook et al., 2012), a fixed CE value may be not suitable. In addition, gas-phase CO<sub>2</sub> will contribute to the CO<sub>2</sub><sup>+</sup> signal and thus influence the mass spectra (Aiken et al., 2007; Aiken et al., 2008). Was the contribution of gas-phase CO<sub>2</sub> to CO<sub>2</sub><sup>+</sup> signal corrected in this study? This information was missed in the manuscript. Furthermore, the authors attributed the difference of SMPS and AMS measurements to shape of the particles. It is unlikely that alone can explain the difference of 5-8 times in concentrations.

3. The authors should give a more detailed discussion of the wall loss corrections of particles. SOA formation and wall loss are competition processes that a significant wall loss would bias the measurements of the SOA in mass loading, composition, and elemental ratios etc... How would the conclusion of high POA/SOA ratio be affected

by wall loss? In addition, the exhaust was transferred through copper tubing and a metal bellows pump (not clear if the system was heated). Did the authors characterize the losses of PM and VOC of this setup? Also, will the metal bellows pump generate particles or VOC? Any data of blank experiments with purified air?

Specific comments:

line 5, page 2: A recent study in HK suggests that COA can be 35% of OA (Lee et al., 2015)

line 19-20, page 1: “after a few hours of chemical aging” is not clear. Can the authors provide information on the OH or O<sub>3</sub> exposures?

line 6-7, page 3: What is the meaning that the cooking particles are the same? Does it mean emission rate of cooking particles?

line 17-18, page 3: A brief introduction of the chamber facility should be given.

line 24-27, page 3: What was the flow rate in the transfer line?

line 28-29, page 3: The RH and T during the experiments should be provided.

line 32-34, page 3: What is the size range of the SMPS? As suggested by the manual of SMPS 3080, the sheath flow should be set to a 10:1 ratio with the aerosol flow. Will the ratio of 5:1 set here influence the measurement of size distribution?

line 17, page 5: How was BC measured in this study? This information was missed in the manuscript.

line 27-28, page 5: The angle  $\theta$  was used for the comparison between different AMS spectra throughout the entire manuscript. It would be useful to explicitly introduce the relationships between  $\theta$  and spectra similarities.

Section 3.2: The authors mentioned that COA emission rates varied due to the different types of meat and cooking procedures. What are the experimental conditions for the studies of Hildemann et al. (1991) and McDonald et al. (2003)? Any suggestions on the influence of meat types and cooking procedures on COA emission rates?

Line 5, page 6: Any evidence that the particles from charbroiling are non-spherical?

Section 3.3: Emissions factors of VOCs were listed here. Any comparison or conclusions? To what extent are they related with the formation of new OA?

line 26, page 6: What was the reaction time for experiment 3?

line 32-36, page 6: It is not convincing that the increase of O:C ratios in experiment 3 was due to the initial presence of ozone. If the increase of O:C ratios are due to the reactions of particles with ozone, the concentration of ozone should have decreased prior to the addition of ozone, which is not reflected in Figure 5b. In addition, as shown in Figure 5a, similar concentration of ozone prior to the addition of ozone was also observed for experiment 2, but no increase of O:C ratios was observed for experiment 2 prior to the addition of ozone. Experiment 2 seems unique with both O:C and H:C increased prior to the addition of ozone. Are there any other explanations for this phenomenon?

line 21-22, page 7: This sentence should be mentioned prior to the description of variations of O<sub>3</sub> and OH concentration. The authors mentioned that similar results were obtained for the rest of the UV illumination experiments. Do the authors mean similar levels or trends of OH concentrations? It is suggested to provide the OH concentrations for all UV experiments.

line 25-28, page 7: What is the definition of new OA here? As the chemical composition of OA changed during aging, should the aged OA be regarded as new OA? This will largely influence the split of POA and SOA. This needs to be clarified. In addition, is there an evidence for the heterogeneous reactions?

line 29-32, page 7: The formation of carbonyls was listed. What is the implication?

line 33-35, page 7: What are the WSOC to OC ratios for the other experiments? The conclusion seems to be based on only one experiment.

line 1-8, page 8: Though detailed PMF analysis was provided in the SI, a brief introduction should be provided here. Please give some explanations on the variations of 2 factors. Did the aged COA factor show some time delay from meal hours?

Section 3.5: This section should be discussed together with the comparison of mass concentrations measured by AMS and SMPS. Also, for comparison, it is better to present the volume mode mobility diameter of particles measured by SMPS.

A table that compares the COA characteristics of this study and those reported in the literature would be useful to readers.

Technical comments:

line 3, page 3: BC and NO<sub>x</sub> are not primary organic aerosol components.

line 13, page 7: “tents” should be “tends”.

Reference:

Lee, B. P., Li, Y.J., Yu, J., Louie, P., and Chan, C.K.: Characteristics of submicron particulate matter at the urban roadside in downtown Hong Kong – overview of 4 months of continuous high-resolution aerosol mass spectrometer (HR-AMS) measurements, *J. Geophysical Research – Atmosphere*, [10.1002/2015JD023311](https://doi.org/10.1002/2015JD023311), 2015.

Aiken, A. C., DeCarlo, P. F., and Jimenez, J. L.: Elemental Analysis of Organic Species with Electron Ionization High-Resolution Mass Spectrometry, *Analytical Chemistry*, 79, 8350-8358, [10.1021/ac071150w](https://doi.org/10.1021/ac071150w), 2007.

Aiken, A. C., DeCarlo, P. F., Kroll, J. H., Worsnop, D. R., Huffman, J. A., Docherty, K. S., Ulbrich, I. M., Mohr, C., Kimmel, J. R., Sueper, D., Sun, Y., Zhang, Q., Trimborn, A., Northway, M., Ziemann, P. J., Canagaratna, M. R., Onasch, T. B., Alfarra, M. R., Prevot, A. S. H., Dommen, J., Duplissy, J., Metzger, A., Baltensperger, U., and Jimenez, J. L.: O/C and OM/OC Ratios of Primary, Secondary, and Ambient Organic Aerosols with High-Resolution Time-of-Flight Aerosol Mass Spectrometry, *Environ Sci Technol*, 42, 4478-4485, [10.1021/es703009q](https://doi.org/10.1021/es703009q), 2008.

Middlebrook, A. M., Bahreini, R., Jimenez, J. L., and Canagaratna, M. R.: Evaluation of Composition-Dependent Collection Efficiencies for the Aerodyne Aerosol Mass Spectrometer using Field Data, *Aerosol Sci Tech*, 46, 258-271, [10.1080/02786826.2011.620041](https://doi.org/10.1080/02786826.2011.620041), 2012.