

Comments:

1. I would like to see a more thorough discussion on how the chemistry in the chamber experiments would be expected to be similar to or different from that occurring in the atmosphere. In particular, I would like to see evaporation of POA, loss of gas-phase species to walls, NO_x concentration, and NO/NO₂ partitioning to be addressed.

We have added a brief discussion about the similarities and differences of our experiments with the atmosphere. The evaporation of the POA is addressed in a separate paper by Louvaris et al. (in preparation) combining isothermal dilution and thermodenuder measurements. For the five chamber experiments the NO_x concentrations were in the range from 1.5 to 8 ppb. The NO₂ to NO ratio ranged from 2 to above 10. There was no detectable decrease of the concentrations of the VOCs measured by the PTR-MS (e.g. loss to the chamber walls) during the characterization periods. The above information has been added to the revised paper.

2. Page 7 line 22: A change in the CE would presumably make the AMS vs. SMPS comparison worse. Please revise and expand this section to provide information on how the suggested factors would influence the comparison and what order of magnitude effect would be expected.

Applying the algorithm of Kostenidou et al (2007), which compares the AMS mass distributions and the SMPS volume distributions assuming spherical particles, we estimated an effective CE around 5. On the other hand, a CE lower than unity would make the comparison even worse. This is similar to the discrepancy between the mass concentrations estimated using the SMPS measurements (assuming spherical particles and density equal to unity) and the filter mass measurements. Explaining this discrepancy would require a density of the COA much higher than unity and the particles to be quite non-spherical. This suggests that the error introduced by assuming spherical particles is of the order of 2-4. This information has been added to the paper.

3. Page 9 lines 21-23: There is a significant body of literature regarding heterogeneous oxidation reactions (e.g., Kroll et al. (2015) and references therein). Are the results presented here consistent with previous works in terms of the O:C and H:C changes observed at the levels of oxidant exposure achieved in these experiments? Given the large body of work on heterogeneous oxidation, a more thorough discussion of this here would be prudent.

The changes observed in Kroll et al. (2015) are qualitatively consistent with our observations, but the corresponding timescales are very different. In our study the changes in both the laboratory and the field take place at OH exposures that are at least one order of magnitude lower than those required in the Kroll et al. (2015) study. However, in our experiments the particles are exposed to ozone also. The results of laboratory studies of oleic acid ozonolysis suggest that the corresponding reactions can take place in as little as minutes (e.g., Morris et al. (2002)), something consistent with the observations here. These suggest that the ozonolysis is probably the most important pathway for the observed changes and not the reaction with OH. We have added a paragraph discussing this important issue.

4. Page 10 lines 8-10: How different in terms of theta were the aged factors from each other? For the aged factor used later in the paper (particularly in Figure 11), was an average aged factor used, or one from a specific experiment? Was there a noticeable difference in between the ozonolysis only factor and those aged with OH?

The aged factors after exposure to UV (Experiments 1, 2, and 4) were similar to each other (theta ranging from 2 to 6 degrees). The corresponding angles between the dark ozonolysis experiment (Experiment 3) and the UV exposure ones were higher ranging from 8 to 14 degrees as the dark ozonolysis factor was less oxidized. The dark ozonolysis factor was between the fresh and UV-aged COA. For the results shown in Figure 11, an average spectrum was used for the fresh factor and the average UV-exposure factor was used for the aged factor. This is now explained in the paper.

5. Page 11 line 7: Here a theta of 13 degrees is defined as “quite similar” whereas on page 6 line 28 a theta of 11-15 is discussed as having “many similarities though they are not the same” and in Sect. 3.4 a change of 15 degrees is discussed as being significant in terms of the changes observed due to ozonolysis. Likewise, the use of “significantly” on page 11 line 27 in describing a change of 15 degrees should be reconsidered. While I recognize that this analysis is somewhat qualitative, it would be beneficial to maintain more consistent descriptions throughout the manuscript.

Part of the inconsistency is due to the comparisons of mass spectra measured in the same experiments and those derived independently by PMF analysis of independent ambient datasets. We have rephrased the corresponding sentences to maintain consistency throughout the manuscript. We have deleted the word “significantly” on page 11 line 27 and just mention the corresponding theta angle.

Technical Corrections:

6. Page 2 line 32: Suggest change to “..indicate that commercial and residential cooking contribute to...”

The proposed change has been included in the revised paper.

7. Page 3 line 3: Suggest change to “...may significantly alter...”
Changed.

8. Page 3 line 7: BC is not a primary organic aerosol component.
The sentence in Page 3, line 7 was rephrased.

9. Page 3 line 23-24: What is the size of the chamber itself?
The proposed information has been included in the revised manuscript.

10. Section 2.1: Please clarify if an OH precursor was used.
No OH precursor was used in any of the experiments. This has been added to section 2.1

11. Page 4 line 8: The voltage difference between the filament and the ion chamber is 70 V.
Changed.

12. Page 4 line 21: “P parameter” should be defined here.
A brief description of the P parameter has been added.

13. Page 4 lines 32-33: Please specify what models of gas monitors.
The gas monitor models have been included in the revised manuscript.

14. Page 6 lines 25-31: It may be useful to indicate why theta is used rather than R² and advantages/ disadvantages.

The advantage of angle theta use for mass spectra comparisons is that it can detect small differences that the correlation coefficient R² cannot. For example, small differences of 1-5 degrees all correspond to an R² =0.99. However, if the difference is quite large (e.g. theta >30 degrees) then R² works equally well. This is now explained in the paper.

15. Please reference the figures in order. Currently Figure 8a is referenced after Figure 1 and before the others (page 7).

The order of the figures has been rearranged in the revised paper.

16. Table 1: Please include the total length of each experiment in the table. In the caption, please indicate that the d-butanol tracer was not added in experiment 1.

Table 1 has been updated with the proposed information.

17. Page 9 line 19: Please consider adding a figure (perhaps to the supplemental material) that show the OA mass change throughout the experiments both with and without wall loss correction.

A figure showing the OA mass change with and without the wall loss correction was added in the revised SI.

18. Page 11 line 17: Given the nature of the analysis, “similarity” rather than “correlation” may be a better word choice.

The proposed change has been included in the revised manuscript.

19. Figure 5: Would it be more appropriate to show the natural log of the PTR-MS signal of butanol normalized to the initial value? Also, please remind the reader that m/z 66 is butanol in the caption.

The caption of Figure 5 now explains that m/z 66 corresponds to d-butanol.

References

Kroll, J. H., Lim, C. Y., Kessler, S. H. and Wilson, K. R.: Heterogeneous oxidation of atmospheric organic aerosol: Kinetics of changes to the amount and oxidation state of particle-phase organic carbon, *J. Phys. Chem. A*, 119, 10767–10783, 2015.

Louvaris, E. E., E. Karnezi, E. Kostenidou, C. Kaltsonoudis, and S. N. Pandis: Estimation of the volatility distribution of organic aerosol combining thermodenuder and isothermal dilution measurements. In preparation.

Morris, J. W., Davidovits, P., Jayne, J. T., Jimenez, J. L., Shi, Q., Kolb, C. E., Worsnop, D. R., Barney, W. S., and Cass G.: Kinetics of submicron oleic acid aerosols with ozone: A novel aerosol mass spectrometric technique, *Geophys. Res. Lett.*, doi: 10.1029/2000GL014692, 2002.