The manuscript 'Photodegradation of Atmospheric Chromophores: Changes in Oxidation State and Photochemical Reactivity' provides results on the photochemical aging of atmospheric aerosols (both ambient PM and laboratory generated POA). The results include OC/EC analysis, parallel factor (PARAFAC) analysis of excitation-emission matrices, and photosensitization of $^1O_2$ with each measured as a function of solar irradiation. The manuscript has been improved but still requires refinement in the writing/presentation and explanation. My comments are outlined below.

Major comments:

1) In the first paragraph of section 3.1, the authors go back and forth between water-soluble and water-insoluble organic matter and WSOC and WISOC. Is there a reason the terminology is different? In Line 184-186, I believe the authors mean to say that the WISOC *decomposes more rapidly* in ambient PM than in POA. If so, rephrase accordingly. In addition, the authors say that ambient PM has been subjected sufficient atmospheric oxidation so that OM is not decomposed, however, the WISOC fraction of ambient PM shows significant attenuation after photolysis. These two aspects seem at odds with one another. More explanation of what the attenuation ratio is would be instructive. This doesn't appear in Sec. 2.4 of the Methods or in Sec. 3.1.

2) In Sec. 3.2, Lines 202-206 are very repetitive stating that the absorbance decreases significantly during photolysis in multiple consecutive sentences. Re-write for clarity. Also in this paragraph, the authors state that the absorbance decay is inconstant and cannot be mathematically analyzed. Do the authors mean that the absorbance decrease cannot be fit to a single exponential decay? Can the authors report a total percent decrease in the absorbance at 350 nm instead?

3) As mentioned above the authors claim that the ambient PM samples have been subjected to sufficient atmospheric oxidation (line 196), however in Figure 5C, these samples are dominated by low oxidation HULIS/C2 (especially compared to the POA samples). How do you reconcile these two observations?

4) In the paragraph starting at Line 274, the authors begin using the term 'light excitation' instead of photolysis or illumination which are used in the figures. It would improve the manuscript to make this terminology consistent. Also, in Line 280-281, what is meant by 'POA has certain oxidability'? Re-phrase whatever concept is trying to be conveyed here.

5) The Implications section is still brief and lacks any reference to previous literature. This section could also be improved by broader interpretation of all the included results. For instance, how do the results in Sec. 3.3 on singlet oxygen generation connect with the results on degree of oxidation in Sec. 3.1 and 3.2, i.e. photolysis increases the degree of oxidation in the aerosol samples which in turn leads to a higher capacity for singlet oxygen formation via photosensitization reactions.

Minor Comments:

Line 2: Change ‘photosensitiveness’ to ‘photosensitivity’ and ‘have’ to ‘has’

Line 10-13: Re-write this sentence: ‘In terms of photochemical reactivity, the triplet state COM *decreases* slightly in ambient particulate matter samples but *increases* in primary organic aerosol (POA) *following photolysis*.

Line 25: Change ‘chemistry’ to ‘chemical’

Line 39-40: Change ‘not complete clear’ to ‘unclear’

Line 58: Change ‘participate’ to ‘participates’

Line 60: Change ‘light conditions’ to ‘solar irradiation’

Line 61: Change ‘induce’ to ‘can generate’

Line 76: Change ‘stated’ to ‘studied’

Line 125: Change ‘could refer to the previous literature’ to ‘has been described previously’

Line 146 Change ‘states’ to ‘state’

Line 229: Change ‘study’ to ‘studied’ and ‘chromophores’ to ‘fluorophores’ to highlight that you are referring to fluorescence here

Line 230: Change ‘, respectively’ to ‘separately’

Line 231: Change ‘so that’ to ‘to’

Line 232: Change ‘chromophores’ to ‘fluorophores’

Line 238: Rewrite as “The composition of the fluorophores changes significantly during the photolysis process.”

Line 260: Change ‘states’ to ‘state’

Line 263 Change ‘not as expected’ to ‘unexpected’

Line 274: Change to ‘further induce singlet oxygen formation’

Line 278: Change ‘is’ to ‘of’