Interactive comment on “Photooxidants from Brown Carbon and Other Chromophores in Illuminated Particle Extracts” by Richie Kaur et al.

Anonymous Referee #3

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This work mainly measured the concentrations of three important photooxidants formed from photoexcitation of brown carbon by collecting ambient particles during heavy residential wood-burning period in winter, extracting them in water, and illuminating the acidified aqueous extracts. The results in aqueous extracts were extrapolated to ambient particle water conditions and compared to the corresponding photooxidants in fog. The main conclusion of this work is that hydroxyl radical in particles had similar levels with fog and cloud drops while singlet oxygen and oxidizing triplet excited sites of organic matters are enhanced in particles. Their results indicate that singlet oxygen and oxidizing triplet excited sites of organic matters formed from the photoexcitation of brown carbon can be important sinks for organic compounds in atmospheric particles. Although there are large uncertainties in the extrapolation to ambient particle water conditions, especially for oxidizing triplet excited sites of organic matters, this work provides the first measurement of singlet oxygen and oxidizing triplet excited sites of organic matters, which affect the lifetime of organic compounds in particle liquid water. The results are very helpful for the science community to improve our understanding of photooxidants and inspire more works for different seasons and locations, and implementing in current atmospheric models. I think this is an interesting and important work, and recommend for acceptance after comments below are addressed:

1. Line 154, please explain “air-saturated”. 2. Line 379, the authors mentioned additional source can be photo-Fenton processes, I am wondering if Fe has been/can be measured in the samples. It would be interesting to compare to Fe data. 3. Line 389-394, suggest adding some literature reviews on the destruction/sinks of OH in this section or in the introduction section. 4. The authors made a couple comparisons between “standard” and “dilute” extracts throughout the manuscript. From the manuscript, the “standard” extraction was based on extracting particles into 1 mL water and the “dilute” was extracting in 2.5 mL water. It is an effect of dilution. It is not clear to me what is the purpose of comparing “standard” and “dilution” conditions. The authors have already studied extensively the effect of dilution using sample 3 in later experiments, so I don’t understand why repetitive comparison were made here or are there additional purposes of comparing “standard” and “dilute” extracts but were not well presented in the manuscript? The authors need to make it clearer. 5. Method section: sample extracts were mixed with photooxidant probes and then illuminated in light. The authors will need to address whether illumination will affect the probes or the products formed from probes and targeted photooxidants. For example, benzene traps OH radicals and form phenol. How does illumination experiments affect the product phenol. Do the authors concern about the photodegradation or photoenhancement of phenol, therefore, resulting in underestimation or overestimation of the OH concentration? Some concerns will also be needed to address for singlet oxygen and triplets cases. 6. Line 339, following the last comment, another concern is the effect of illumination on the light absorbance of brown carbon. For example, a recent study by Wong et al. 2017
EST (Changes in Light Absorptivity of Molecular Weight Separated Brown Carbon Due to Photolytic Aging) showed that light Absorptivity of brown carbon changes due to photolytic aging. Please discuss how change of absorptivity affects the conclusions in this work.