Reviewer #2

We would like to thank the Reviewers for their constructive feedback. The reviewer comments are reproduced below in black, and our responses are provided below each comment in blue. We will also be uploading a track-changed version of the manuscript along with all responses as required by ACP.

1) The extraction procedure in the first paragraph of section 2.2 requires further elaboration. What is the rationale behind using the 3 organic solvents in these proportions? Is that based on an already established protocol? Have you verified that none of the BrC precipitates out of the water+DMSO mixture after evaporating the organic solvents? Why are there different extraction protocols for the fresh (water+DMSO) and photolyzed (only DMSO) particles?

An earlier publication (below) established that this organic solvent mixture resulted in the highest light absorption over the near UV and visible range, presumably due to a higher extraction efficiency. This is represented in Figure 1 of the paper. This paper is now cited.

Lin, P., Bluvshtein, N., Rudich, Y., Nizkorodov, S., Laskin, J., & Laskin, A. (2017). Molecular Chemistry of Atmospheric Brown Carbon Inferred from a Nationwide Biomass-Burning Event. Environmental Science & Technology, 51(20), 11561–11570. https://doi.org/10.1021/acs.est.7b02276

By visual inspection, there is not any precipitate from the water/DMSO mixture. However, there could be small, insoluble particles in the solution which we have not been able to see scattering from since the volume is very small (150 microliters). This is now clarified in the paper on page 6.

For the extracts of photolyzed extracts, the volume is much smaller at 30 microliters, and because many of the chromophores seem to dissolve better in DMSO than water, we simplified it by just adding DMSO. This does not affect the chromatography or the quality or the photodiode array spectra.

2) Line 235: Along the same lines, why is the extraction procedure for UV-vis different from HPLC/PDA/HRMS?

The extraction solvent mixture was altered depending on their advantages and disadvantages for the application: HPLC/PDA/HRMS or UV-Vis spectroscopy. Dichloromethane was avoided in the overall BrC photolysis experiments (UV-Vis experiments) and methanol was used instead. Dichloromethane absorbs strongly at <240 nm, and could interfere in the measurement of BrC absorption at near-UV wavelengths. In addition, from a safety standpoint, DCM is not a convenient solvent for absorption spectroscopy since it needs to be used in the hood due it its high volatility. Regardless of the solvent mixture used, it is impossible to extract all the material from filter with any solvent combination, and this is why the transmission spectroscopy measurements of the entire filter were done. The decay of absorption of visible wavelengths was consistent across both measurements (solution versus filter).

We now have explained the origins of the extraction protocol on page 6. An additional sentence on page 8 discusses why methanol was used in place of DCM.

3) The binning of the chromophores into Major, Intermediate, and Weak is informative, but can be confusing. I think it is helpful to stress that these categories involve the combined effects of abundance and absorption efficiency. For instance, an abundant chromophore with moderate

absorption efficiency can exist in the same bin as a less abundant chromophore with high absorption efficiency.

This is a good point. Our approach in this paper is to emphasize the observed contribution to the overall absorption by individual chromophores (rather than their relative concentration). Species such as nodakenetin ($C_{14}H_{14}O_4$) are a major contributor to the observed absorption for many of the fires. However we have not run standards to ascertain the emission factor for each compound and cannot determine the relative abundances of any of these compounds. This was the focus of Jen et al., 2018, which quantified the emission of particle phase compounds from the same fires, publishing the % contribution from each of the compound classes. To emphasize this point, we added the following sentence on page 9.

"Abundance and absorption cross section of BrC chromophores both factor into their assigned absorbance bin, as absorbance was not mass normalized with standards. It is possible that the chromophores labelled as "M" are present in small concentrations but have large absorption coefficient."

4) In addition to integrating the DPA signal over 300nm – 700 nm, can the authors also report wavelength-dependent information? Are there significant differences in AAE across the different chromophores? If yes, integrating absorption over say 4 sections (UV, short visible, mid visible, long visible) can provide interesting information. For example, a chromophore under category W but with say a relatively large AAE might be an I or an M in the UV range.

In general, BrC chromophores in this study are most absorbing in the UV and near UV, with some extending their absorption into the visible. While many have features in the near UV and visible with different λ_{max} values, their relative integrated absorbance does not change all that much depending on the wavelength range (200-700 nm versus 300-700 nm), although many chromophores are excluded if only including visible wavelengths (400-700 nm). We have included absorption spectra in Tables 1 and 2 so that readers can check which chromophores are important in certain wavelength ranges (such as the long visible).