

## **Reviewer #1**

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.

### Major comments:

With regards to calculating aging times: The authors integrated over the spectral flux for the two types of lights and normalized this to the spectral flux of the sun. However, shining light onto a particle collected onto a filter is different than shining a light on an aerosol particle. The filter will absorb and reflect light back through the collected particle, which will alter the amount of radiation the particles experiences compared to aerosol particles up to double the amount of radiation. Do the authors know how the filter will alter calculated photochemical lifetimes?

This is an excellent point, which we neglected to explain in this paper. Presser et al. (2014) measures the absorption enhancement due to scattering by different kinds of filters coated with nigrosine dye. They found that the absorption enhancement was highest for the Teflon filters, and was highly dependent upon the mass loading and temperature, and perhaps even pore size. This is an additional source of error in our lifetimes since we are not able to quantify the absorption enhancement specifically for our filters. We now clarify in the paper that these lifetimes of BrC absorption and chromophores are lower limits. It is now mentioned three times in the paper, once in the Experimental section and twice in the Results and Discussion section (Pages 9, 12, 13).

Page 12, line 366: The authors comment how the same chromophore exhibited different lifetimes between fuels. Judging from figure 5, the range of lifetimes is within a factor of 2-3 (0.4-1.2 days). The authors provide several reasonable explanations on why the wide range of lifetimes for the same chromophore (or compounds that elute at the same time). However, the authors in the previous paragraph note that brown carbon chromophores from chamise burns have shorter lifetimes than lodgepole pine burns. Can the authors comment on the uncertainty range for these lifetimes as it seems like the variation for just one chromophore is enough to explain and differences in specific chromophore lifetimes observed between fuels.

This comment must have been triggered by our lack of error bars in the initial submission. We have added error bars for the lifetimes provided in the paper (Figure 5 and Table 4). Error bars were calculated from the standard error of the linear trendline's slope, which is the first order rate constant. This error only accounts for the uncertainty in describing the measurements with the model. There are uncertainties in the measurement method that cannot necessarily be quantified, such as differences in extraction efficiency between photolyzed and unphotolyzed parts of the same filter and the final concentration volume of HPLC/PDA/HRMS samples.

### Minor comments:

Page 2, line 47: "Wildfires continue to. . ." This sentence semi-repeats previously stated information.

We agree that the sentence is somewhat repetitive and disrupts the flow of the paragraph. It was deleted.

Page 2, line 59: "large effect on radiative forcing" In which direction?

It is now clarified in the paper with “positive.”

Page 2, line 65: “broader range” broader range than what?

We mean broad range in terms of the diversity of molecules and combustion sources which can produce different molecules. We hope it is clearer now.

Page 4, line 123: “irradiated BBOA water extracts. . .” The phrasing of these sentences a bit confusing as it seems like the solution just randomly lost WSOC.

Thank you, we clarified that this is in response to irradiation of the extract.

Page 4, line 126: “produced from Alaskan peat” produced from burning Alaskan peat

Clarified.

Page 5, line 138: the semi colon is not used properly

We replaced the semicolon with a comma in the revised sentence.

Page 6, line 173: How long were the filters stored for before analysis? If it was a long time (months later), do the authors know/guess how these chromophores degraded with time?

In the revised manuscript, we added that the BrC chromophore analysis was done no more than 2 months later. Since the samples were always frozen, we do not expect BrC chromophores to decay on this timescale (although no one has ever verified the chemistry occurring on frozen filters!). The partitioning of semi-volatile components may be different from the fresh emissions, but we are not as concerned with comprehensive particle composition in this paper.

Page 9, line 252: For the structures of compounds given in Table 1, did the authors run standards to try to better confirm the identities/structures of the reported compounds? By reference spectrum, do the authors mean spectra from running standard compounds?

“Reference” was corrected to “standard.” Standards were run for select compounds based on availability. The caption in Table 1 was clarified to show that the standard spectrum is in blue.

Page 14 line 418: Please provide another sentence or phrase explaining why photodegradation occurs more rapidly in solution.

The sentence has been added to and now reads:

“However, in Lin et al. (2016) BBOA was extracted and irradiated in solution where photodegradation could occur more rapidly due to molecular diffusion (Lignell et al., 2014).”

Page 14 line 419: no comma after campaign

Done.

Page 14, line 420: What does AAE stand for?

Thanks. This was corrected.

Figure 1 and 2: Since there are not too many lines, please make the lines distinguishable for black and white printers.

Thanks for the suggestion. With four traces, the authors prefer colors rather than using dash/grayscale color variations.

Figure 2: Maybe label on the graph the two compounds.

Thanks, we took your suggestion.

Figure 3 and 4: There appears to be a bracket-shaped line border around these figures. Can this be removed? Also, caption says photolysis but which type of photolysis?

These lines seem to appear in PDF conversion. We will work with the publisher to edit them out from the final version. Thanks, we have clarified that this is 300 nm photolysis.

Figure 5: Along with the comment from before, it would be useful to have uncertainty bars on this figure so the reader knows what constitutes a pattern or not.

Please see the above comment. In short, we did add uncertainty bars.

Table 1: Why do the peak # stop after 43?

Thanks, this was corrected.

Table 3: What does "Figured used" column mean? Is this important information?

The title of the column is now "Sample used in figure #" and it was moved to the SI (now Table S2).

Table 4: The caption says irradiation was done for condensed phase, filter, and all samples. This is confusing as the columns only reference condensed phase. Overall, the column headings are confusing. Should average not be capitalized? And averaged?

The column headings were simplified/changed to "BrC absorption lifetime averaged LA (equivalent days)."

Supporting Information:

Figure S1: Please make this graph more readable. Which equation goes to which line? Which lines describes which points? There appears to be only two data points on the condensed phase line (not counting 0,0). Why were more points not taken? Seems like 0,0 is dominating the trend line which makes the measured points seem very off. Also please add in the text that this method was used for both types of aging experiments (specific chromophores and overall BrC) unless this is not the case.

The trend lines are very similar, and we agree it is difficult to tell which is which, although the  $R^2$  is a good indicator that it's the solution phase measurement (since there's only two points). We agree that more points should have been taken to be more confident in the trend. However, it is important that the trend line go through 0,0 and it is important to keep in mind that the photolysis will likely not follow a first order decay process perfectly, and some variation is expected.

The following is said in the text (SI p3). We have also clarified the title of the section in the SI to "Calculation of the estimated BrC absorption lifetime and individual BrC chromophore lifetime"

"This procedure is used for individual chromophores as well as overall BrC absorbance."

Relating to the previous comment, there seems to be pretty high uncertainty in estimating lifetimes as this Figure S1 shows 1-2 data points and a linear regression. Please add a significant discussion on uncertainty bounds.

Please see the above comment about added error bars.

Is condensed phase filter-based? In the main text (page8 line 239), you refer to both filter-based and condensed phase. Could you please clarify which term refers to what?

We now explicitly state “filter-based” when contrasted with “solution phase” absorption spectroscopy measurements. Condensed phase is still used in the paper, but only used to describe the particulate matter photolysis.