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Atmospheric Chemistry and Physics
Discussions

Interactive comment on "Light absorption by polar and non-polar aerosol compounds from laboratory biomass combustion" *by* Deep Sengupta et al.

Deep Sengupta et al.

vera.samburova@dri.edu

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RESPONSES TO REVIEWER 1

This study examined the optical and chemical properties of fresh and aged aerosols from laboratory biomass burning (BB). The effect of polarity, pH, molecular weight, biomass fuel type, and combustion conditions [flaming versus smoldering] on the aerosol optical properties were reported. The aged aerosols were produced in an oxidation flow reactor (OFR). The optical and chemical properties of the aerosols were measured using UV-Visible spectroscopy and high-resolution LC-MS, respectively. A spectrally integrated mass absorption efficiency was calculated, and Van Krevelen diagrams were used to visualize chemical information about the extent of oxidation. The

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study found that non-polar matter in the organic aerosols was more absorbing than the polar fraction; although, water soluble matter in the aged aerosol was more absorbing during flaming. Low pH aerosol was less absorbing than at neutral pH. The study also provided convincing evidence that the NOx formed during flaming combustion yields organic nitrogen compounds that are strongly absorbing at shorter wavelengths. This is a useful, well-done study that is within the ACP scope. Biomass burning is an important source of organic aerosol in the atmosphere, and its optical and chemical properties are key to understanding the effects of aerosols on climate, air quality, and human health. The results presented are novel in that aerosol optical properties are measured after being fractionated in-bulk using multiple chemical methods. The investigation of fresh versus aged aerosol also produced an interesting contrast. The examination of peat is something that is in its infancy and more peat burning studies are needed owing to the increasing potential for peat mega-fires forming globally. Mostly, the chemical and optical data synched together nicely and evidence-based conclusions are given. Although, with so many variables, sometimes it was difficult to track the rationale underlying an experiment and the obtained results. The comments below are meant to produce further clarification regarding the experimental results. The paper may also benefit from a forward-looking statement that describes specifically, where some of these results may be useful or applied. Such a statement can come in the summary or at the end of the introduction. After these minor revisions, the paper merits publication.

AC: We thank the reviewer for his/her comments and recommendations. We address comments individually below. Approximate line numbers corresponding to the edited (with markup) manuscript is provided.

Comments: 1. P1, line 10: It sounds like BrC is the only light-absorbing component in BB aerosols. This opening sentence should be re-phrased for clarity sakes.

AC: The sentence was rephrased.

2. P5, line 23: Please describe the pre-firing technique (i.e., add a temp. and time). AC:

The “pre-firing” conditions (temperature and time) for 47-mm quartz filters are added to the text.

3. P6, line 12: “and” should be or.

AC: “and” was changed to “or”

4. P7, lines 13-14: It would be a good idea to tell us how much time it took to complete each type of burn and provide a better idea of the sample time weighting.

AC: Thank you for the recommendation. The following sentence was added:“The duration of smoldering combustion was 58ïĆś27 min on average, whereas the average duration for flaming combustion was 22ïĆś8 min.”

5. P7, lines 22-25: It’s not clear why the NO2/m was handled this way. Is the assumption that the fuel burned the same over the entire test even though sampling time was limited?

AC: We agree. NO2/m was not used in the present work (only NO2 max values were presented, in ppb). These sentences were removed from the text.

6. P.8, Section 2.4: Where is the extraction efficiency calculation? It would be useful to see what fraction of aerosol was removed from the filter using each solvent?

AC: We thank the reviewer for bringing up this important point. We also feel a calculation of extraction efficiency/percent recovery would be useful as we are comparing absorbance values from water and hexane extracts. Sonication is a standard technique for extraction of water soluble aerosol components from filters for which almost 100% efficiency is assumed (Fang et al., 2015; Gao et al., 2017; Kiss et al., 2002; Samburova et al., 2005). In our study, sonication was performed twice to ensure maximum recovery. An accelerated solvent extractor (ASE) was used to recover hexane soluble fraction from the filter. The ASE uses high solvent pressure and temperature to ensure maximum recovery. A statement to this effect was added to the experimental section (section 2.4, Page 8, Line 25).

7. P10, lines 7-11: This sentence needs to be clearer. AC: The paragraph was revised.

8. P11, lines 3-9: Same thing, this sentence requires clarification. AC: The paragraph was re-written.

9. General comment: It would be beneficial if there was some additional discussion about what is expected to happen during the aging in the OFR. Was the OFR producing new particles? Or was it changing the existing particles via heterogeneous chemistry? Or both? Which of these processes/mechanisms is likely to dominate? What are the particle loss characteristics of this specific OFR? With the knowledge that BB fuel types produced different aerosol types, is one aerosol type more susceptible to loss than another? More information that helps us assess these results should be added here.

AC: We appreciate reviewer's concern about more detailed description about OFR characterization and fate of organics during aging. Detailed OFR characterization and discussion of various mechanism occurring in the OFR and their timescales can be found in another manuscript associated with this work (Bhattarai et al., 2018) (under review). We have added a sentence in section 2.3, page 7, lines 10-12.

To address reviewer's questions we will briefly summarize the main outcomes of Bhattarai et al., (2018) paper. We found that a wall loss of low-volatile organic compounds (LVOC) and semi-volatile organic compounds (SVOC) is negligible (0.1%) in the OFR. However, about 25% of primary organic aerosol mass is lost due to fragmentation reactions. An approximately equal amount of secondary products condenses onto primary particles. We did observe new particle formation in the OFR.

Particle loss in the accumulation size range was found to be less than 10%. Since the aerosol mass was concentrated in this size range, we do not expect such losses to be significant for our absorbance measurements which are mass-based. Diffusional particle losses depend only on particle size and not on chemical properties. Thus, we do not expect any differences in particle losses among different aerosols tested in our study.

10. P18, lines 3-4: Revise this sentence for clarity. AC: The sentence was rephrased.

11. P18, lines 1-2: Not 100% convinced about this. It may simply be that one is measuring less of something thus the uncertainty is a larger fraction of the measurement. AC: We agree with the reviewer and the sentence is removed from the text.

12. P18, lines 14-17: The fuels could have been tested for their N content. Are those data available? AC: Thank you for a valuable suggestion. We plan to measure the nitrogen content of the fuels in future. Unfortunately, these data are in not available at the present time.

13. Figure 5: Mention in the caption that this set of plots is for WSOC. AC: We thank the reviewer for pointing this out. All ultrahigh resolution mass spectrometry results demonstrated in this work are from water soluble organic carbon (WSOC). We have added WSOC in the Figure 5 caption as per suggestion.

14. P22, lines 13-15: Regarding DBEs, it sounds like that bias is built into the measurement via the ionization chamber. Should that be further addressed?

AC: This is a well-known bias with electrospray ionization. In the negative ion mode, only the molecules with an active H (e.g., carboxyl groups) can be deprotonated. In this case, we wanted to see which new species were formed by oxidation, so the bias is quite useful. To clarify this, the following sentence was added to page 13 line 11.

15. P27, lines 11-12: Not sure Fig. 8 supports this observation in general for both aged and fresh aerosols. Please clarify.

AC: The statement on page 27 holds true only for fresh aerosols. This point is clarified in the revised manuscript as per reviewer's suggestion

16. P28, lines 17-18: Does it mean anything that smoldering biomass emits more per unit mass of fuel burned? We're getting deep into the manuscript now, and it may be

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good to remind the reader that everything is normalized to weight or volume or. . .... If it is?

AC: In page 28, line 17-18 Our results showed that the secondary organic aerosols (SOA) produced by OFR oxidation (OFR-aged) of BB aerosols from flaming combustion type fuel have larger light absorption efficiencies. In contrast, the OFR-aged BB emission generated by smoldering combustion of peat fuels absorb less than fresh BB emissions from the same fuels. The absorption efficiency was normalized by mass of OC. The units' explanation was added (Figure 8 capture and P. 28, line 17)

17. Figure 10: Why was no attempt made to examine the hexane soluble extracts? May want to briefly raise this point in the Experimental section or while or after discussing the virtues of this result.

AC: Oxidized (ionized) molecules were not expected to be observed in the hexane extracts. Our intention was to inform the water-soluble OM composition since it's the most likely fraction to contain the oxidized components that are amenable to ESI. In future studies, we can use APPI (Atmospheric Pressure Photoionization) to further explore the composition, however, that was deemed beyond the scope of the current project.

18. P25, lines 4-18: This discussion starts earlier at page 21 (lines 14-15), but doesn't really take off until here. AC: We apologize, but we are confused regarding this comment and are not sure what the reviewer is referring to.

19. Figure 11: This is a nice summarizing figure. Although, why are only select experiments from the current study shown here? Consider showing all the experiments.

AC: Thank you! We also think that imaginary refractive indices for all fuel extracts (polar and non-polar) are important to present with the data reported from the previous works. However, when we tried to include all data sets for all fuels, the figure became too busy and hard to read. For this reason, we selected only two fuels and refractive indices for water and total extracts for the comparison. In the Supplementary Material, we presented the imaginary refractive indices of water (Fig. S5) and hexane (Fig. S6) extracts from all fuels and for both fresh and aged aerosols.

Please also note the supplement to this comment:
https://www.atmos-chem-phys-discuss.net/acp-2018-161/acp-2018-161-AC1-supplement.pdf

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