

## Review for amt-2019-5 “Two-wavelength thermo-optical determination of Light Absorbing Carbon in atmospheric aerosols” by Massabò et. al.

The manuscript reported a modified carbon analyzer with dual-wavelength configuration for the determination of Brown Carbon. Multiwavelength thermal-optical analysis had been reported using the DRI carbon analyzer (Chen et al., 2015; Chow et al., 2015; Chow et al., 2018), but multiwavelength applications on the Sunset analyzer remain limited (Hadley et al., 2008). In that sense, this study has the merit from the instrumental perspective. However, part of the data analysis suffered from overinterpretation, thus, revisions are needed.

### General comments:

- 1) How this study can be beneficial to the carbonaceous aerosol research community? From the instrumental perspective, there is already a multiwavelength carbon analyzer that is commercially available (DRI2015), as noted by the authors. The modification described in this study might not be easy to be adopted and implemented by other research groups. The authors need to elaborate how this setup can be implemented by other researchers.
- 2) Introduction. Beside primary BrC from biomass burning, the secondarily formed BrC should be mentioned.
- 3) The current modification only allows one laser to be used at each time, that means all samples need to be analyzed twice. As noted by the authors, the change of laser and PD require alignment to optimize the laser signal. Since laser and PD change would introduce further uncertainties into the OC/EC analysis, this point should be mentioned. Did the authors quantify the uncertainties in OC and EC determination that introduced by the change of laser and PD? For example, what's the standard deviations of OC and EC from multiple analysis for the same sample (identical laser and PD with mount-unmount cycles scenario vs. no laser and PD change scenario)? The authors are encouraged to provide a estimation of uncertainty introduced.
- 4) The  $MAC_{BrC}$  reported in this study ( $9.8 \text{ m}^2\text{g}^{-1}$  @635 nm and  $23 \text{ m}^2\text{g}^{-1}$  @405 nm) seems to be one magnitude higher than the literatures values. An example is shown below. The following table was adopted from (Updyke et al., 2012). The author argued the difference is due to the operative defined BrC mass used in this study. It should be noted that literature studies applied different technical approaches for  $MAC_{BrC}$  determination as well, but most studies reported a  $MAC_{BrC} < 1 \text{ m}^2\text{g}^{-1}$ . The author should explain why their results are significantly different from previous studies.

Values of MAC in  $\text{m}^2 \text{g}^{-1}$  from representative field and laboratory studies of organic aerosols measured at or near 500 nm. The cited MAC values explicitly remove contributions from black carbon. With the exception of this work, all data cited in this table likely correspond to primary sources of brown carbon.

Sample (campaign)	$\lambda$ (nm)	MAC ( $\text{m}^2 \text{g}^{-1}$ )	Reference
Brown carbon produced by aging SOA with 100 ppb $\text{NH}_3$ (lab)	500	0.001–0.1	This work
“Tar balls” from smoldering combustion of wood; brown carbon contribution (lab)	532	0.01–0.07 (calculated from $k = 0.0005$ – $0.003$ )	(Chakrabarty et al., 2010)
HUmic-Like Substances (HULIS) extracted from filter samples from various sites in Europe	532	0.07–1 (calculated from $k = 0.003$ – $0.05$ )	(Dinar et al., 2008)
Methanol extracts from wood combustion particles (lab)	500	0.1–0.5 (estimated from the graphs)	(Chen and Bond, 2010)
Refractory organic carbon from biomass burning in North America (INTEX/ICARTT)	470 530	0.6 0.1	(Clarke et al., 2007)
“Tar balls” in North America (YACS)	632	0.4 (calculated from reported $k = 0.02$ )	(Hand et al., 2005)
Brown carbon in pollution plumes from Asia (CAPMEX)	532	0–1	(Flowers et al., 2010)
Brown carbon in particles collected in Asia (EAST-AIRE)	520	0.6	(Yang et al., 2009)
Acetone extracts from biomass burning aerosols in Africa (SAFARI 2010)	500	0.9	(Kirchstetter et al., 2004)
Amorphous carbon spheres from biomass burning (ACE Asia)	550	4	(Alexander et al., 2008)

- 5) Line 277. Regarding the BrC mass determination using the method reported in Massabò et al. (2016), did the author considered laser-temperature correction (Jung et al., 2011) ? Seen from Fig 5 in Massabò et al. (2016), the laser signal keep increasing during the CH<sub>4</sub> stage, implying that laser-temperature correction was likely not performed. If that's case, the BrC mass should be re-calculated. In addition, even if the laser-temperature correction is accounted, the laser uncertainty is simply too high for BrC mass determination. Please specify the limit of quantification (LOQ) for OC in the OC/EC analysis. The reviewer feels that LOQ<sub>OC</sub> would be likely very close to the level of BrC reported in this study (0.005 – 0.14 µgC m<sup>-3</sup>). If so, BrC reported using this approach is overinterpretation of the data.
- 6) The BrC determination approach described in Massabò et al. (2016) lacks physical meanings. The OC/EC split by the laser signal in the thermal optical analysis depends on two assumptions: (i) pyrolyzed organic carbon evolved before native EC during the oxygen stage. (ii) pyrolyzed organic carbon and native EC have the same MAC. However, both of these assumptions had been proved invalid (Yang and Yu, 2002; Yu et al., 2002; Subramanian et al., 2006). The approach that author used is a paradox: On one hand the authors report a MAC<sub>BrC</sub> that is larger than MAC<sub>EC</sub>. On the other hand, the laser correction process itself is based on the assumption that MAC<sub>BrC</sub>=MAC<sub>EC</sub>=MAC<sub>POC</sub>. In that sense, the carbon fraction corresponding to the different laser split time cannot be considered as BrC mass.
- 7) The authors are encouraged to check the b<sub>abs,BCff</sub> vs. levo scatter plot. If the R<sup>2</sup>(b<sub>abs,BCff</sub> vs. levo) is significantly lower than the R<sup>2</sup>(b<sub>abs,BrC</sub> vs. levo), that would be a useful evidence to confirm a successful split of b<sub>abs</sub> into BrC, BC<sub>WB</sub> and BC<sub>ff</sub>.

#### Technical comments:

- 1) The figure quality needs to be improved. For example, for comparison of the same quantity/parameter, the X and Y range should be the same and the aspect ratio of the plot should be 1:1.
- 2) Figure 1. Please label the laser wavelength on the photo directly for easy reference.
- 3) Figure 2-7. The font size is too small for the text in these figures. Please adjust accordingly.
- 4) Figure 5 caption. “WW and FF stand for Fossil Fuel and Wood Burning, respectively.” Should be “FF and WB stand for Fossil Fuel and Wood Burning, respectively”
- 5) Line 245. “and biomass burning (WB)” should be wood burning?

#### References

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