

**Title of the paper:** Evaluation of the carbon content of aerosols from the burning of biomass in the Brazilian Amazon using thermal, optical and thermal-optical analysis methods  
**Soto-Garcia, L et al.**

**Answer to Referee #2**  
**General comments:**

**1. Referee#2: The methods used by the authors are largely correct, yet the reviewer is not convinced that this paper would add much to our understanding on aerosol carbon chemistry or methodology.**

Answer: For biomass burning samples, there is no real sharp boundary for differentiating OC from EC<sub>a</sub>/BC<sub>e</sub>, due to the presence of OC material that is highly refractory and optically absorbing. Size-resolved EC/BC and OC concentrations are needed to improve our understanding of the lifetime of these species as well as of the optical properties of biomass burning and, therefore, to improve climate predictions, particularly since EC/BC climatic effects strongly depend on its physical and chemical properties. This manuscript provides valuable information in this regard. Our principal objective was to evaluate different methods for determining the carbonaceous material in biomass burning samples so that we could eventually present and discuss the size distributions of the species that composed this fraction (i.e., EC<sub>a</sub>/BC<sub>e</sub> and OC). We used the following methods: TOA, TOT, TOR, carbon monitor, LTM, aethalometer, together with water extraction procedures, to evaluate the results for EC<sub>a</sub> and BC<sub>e</sub> (Sections 3.1-3.2). This allowed us to observe that after the water extraction, the charring diminished significantly, and EC<sub>w</sub> results were similar to the BC<sub>e</sub> from the optical method LTM at 880 nm (using a specific absorption cross section of 16.6 m<sup>2</sup> g<sup>-1</sup>), and to the EC<sub>a</sub> from the TOR. Also, from the results of the Ångström exponents,  $\alpha_x$ , obtained from the optical data of the TOA after the extraction process (Section 3.2), we observed that probably brown carbon (C<sub>brown</sub>) is interfering with the EC<sub>a</sub>/BC<sub>e</sub> signal. After, estimating the best EC<sub>a</sub>/BC<sub>e</sub> we produced the size distributions of the carbonaceous material (OC and BC<sub>e</sub>). These results are indeed an important contribution to our community since we propose different methods to determine the best approach to determine EC<sub>a</sub>/BC<sub>e</sub> in biomass burning samples from the Amazon Basin, and at the same time we highlight what the interferences of the EC<sub>a</sub>/BC<sub>e</sub> results can be. We have added this information (importance of size distribution of the carbonaceous material) to the introduction.

This manuscript also provides valuable information regarding the use of quartz filters with the DLPI. Size-resolved carbonaceous particles were collected on quartz fiber filters placed on the DLPI in order to be able to analyze them using the thermal and optical methods used here. In this manuscript we provided an estimate of the shift found in the TC size distributions due to the use of quartz instead of aluminum substrates.

**2. Referee#2: In particular, the collection of particles on quartz filters in the Dekati impactor is questionable, since these sampling devices are originally not designed for that (despite some efforts to apply filter substrates in impactors). I understand that the application of TOR/TOT analyses and LTM would require quartz instead of quartz filters, but are the results really meaningful? There**

**are so many problems with filter substrates in themselves (artefacts), why add other potential errors to the already problematic aerosol sampling?**

Answer: Quartz fiber filters are needed for the analysis of the carbonaceous fraction using LTM, TOR and TOT as the referee mentioned. These filters have been used before in the determination of the size-resolved carbonaceous fraction using impactors (e.g. Saarikoski et al., 2008; Herckes et al., 2006). We know they introduce biases in the size distributions when impactors such as the DLPI are used. For that reason we devoted several sections (Section 3.1.3, 3.1.4, and 3.4) to explain and to estimate the errors in the determination of the DLPI size distributions (comparison of TC concentration collected on the aluminum substrates vs. the TC collected on the quartz fiber filter, now Figure 7). The shift in the size distribution when using quartz filters instead of aluminum substrates is very useful information for other researchers who have an interest in using the DLPI (or SDI; see Saarikoski et al., 2008) with quartz filters.

**3. Referee#2: The most valuable part of the work is to demonstrate to what extent apparent black carbon (charring of organic compounds and partly brown carbon) contributes to measured BC concentrations.**

Answer: We are glad that the reviewer appreciates the merit of this part of our work. We feel, however, that other aspects, such as the size distribution of  $EC_a/BC_e$  and the thorough intercomparison of chemical and optical analytical techniques also are valuable contributions.

Also, it is certainly possible that charring products of OC and refractory OC coevolve with EC during thermal analysis, in which case they contribute to the  $EC_a$ . For the determination of charring  $EC_a/BC_e$  we need to measure pure BC and pure char (Hadley et al., 2008), neither of which were measured here. Regarding the estimation of brown carbon from spectral absorption we must know the mass absorption cross-section of the brown carbon, which we do not know well. This topic certainly needs more study.

**4. Referee#2: It was interesting to see how the Angstrom exponent was reduced after water extraction: it was an indication that brown carbon absorption was also significant in the UV range of spectrum, and not only extensive charring distorted the light absorption signal. Perhaps some estimates may have been given on the relative importance of the two as a function of wavelength.**

Answer: The difference in OC absorption before and after extraction is an indication that brown carbon contributes to aerosol light absorption. It is not clear that all of the extracted carbon is light absorbing, and it is not clear that all of the light absorbing OC is extracted, so one cannot estimate the OC mass with a great degree of certainty from the extracted mass. To estimate the mass of brown carbon from spectral absorption measurements, one must know the mass absorption cross-section of the brown carbon, and unfortunately this value is not known with sufficient accuracy.

**5. Referee#2: Minor comment: Page 12881 Line 2: In biomass smoke, cloud processing may also happen in a persistent smoke layer, which is in itself a type of cloud. So there is no need to assume extensive ageing to explain the occurrence of the droplet mode biomass burning aerosol.**

Answer: Based on the referee's suggestion we deleted part of that sentence and now it reads as follows: "The submicron size range we are reporting here is usually called the droplet mode (0.4 – 0.6  $\mu\text{m}$ )."