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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Evaluation of the carbon content of aerosols from the burning of biomass in the Brazilian Amazon using thermal, optical and thermal-optical analysis methods" *by* L. L. Soto-García et al.

Anonymous Referee #1

Received and published: 6 July 2010

General comments:

The manuscript presents bulk and size-segregated carbonaceous aerosol composition measurements for filters collected in Brazil during the biomass burning-impacted dry season. It focuses on characterizing the organic/elemental carbon composition and light absorption properties, in some cases as a function of size. Many of the observations are consistent with a number of previous studies, including other measurements in the Amazonia region, including bulk mass concentrations, the distribution of elemental and organic carbon mass, and the spectral dependence of absorption. The manuscript also compares the results from a number of EC/BC measurement methods, including the commonly used thermal optical reflectance, thermal optical transmittance, and aethalometer methods. The topic of the manuscript is relevant to ACP and the paper is generally well written. Unfortunately the analysis in the paper is somewhat limited, and it seems to lack a clear focus. At times it feels like a methods inter-comparison paper that discusses differences between the different EC and BC measurement techniques in detail, but at other times focuses more on the novel size-segregated OC/EC and light absorption measurements. Both topics are important and worth detailed study, but at present the manuscript does not accomplish either of these objectives particularly well.

The EC/BC methods inter-comparison aspect of the paper requires more attention to the role of sampling artifacts on the interpretation and additional comparisons to previous inter-comparison exercises. Additional attention should be paid to the TOT and TOR results (e.g., how large were pyrolysis corrections in each case, how do these values compare to the other methods, were there day/night differences?).

The size-segregated results would also benefit for a more in depth analysis. These are, to my knowledge, the first, or at least among the first, size-dependent measurements of the wavelength dependence of absorption, yet the results in Table 4 are barely discussed in the text. Organic carbon and elemental carbon data are presented as a function of size, but it would be interesting to include a discussion of changes in the EC/TC ratio as a function of size. Can this be linked to changes in the spectral absorption dependence? What broader implications would these results have on optical properties and/or CCN activity of the emissions?

The organization of the paper could also use improvement. It isn't clear why the topics described in sections 4 and 5 are worthy of their own sections while a large number of the other results are combined into section 3. The manuscript tends to jump between the two topics described above, making it somewhat difficult to follow. For example, the mass and TC distributions are presented in Figure 1, but the reader has to wait until Figure 9 to see the organic and elemental carbon results. If the two topics can be better organized and expanded the significance of the findings would be clear, or if the focus of the paper was shifted to either topic, the manuscript would suitable for publication in ACP.

Specific comments:

page (line)

12861 (16): Dp is not defined.

Introduction

The introduction provides a detailed review of previous biomass burning aerosol composition measurements in the Amazon, including size-segregated results that can be linked to aerosol hygroscopicity and CCN activity. A significant amount of the manuscript justifies previous organic composition (e.g., WSOC) measurements, but there is little motivation provided for size-segregated OC/EC measurements. The introduction itself states that existing measurements were sufficient to predict aerosol hygroscopic properties and CCN ability over the Amazon (12864, lines 5-6), so what additional benefits do the observations presented in this study bring? This could be addressed by a discussion that gives some of the benefits size-segregated OC/EC measurements specifically provide (e.g., better constrain EC lifetime, improve treatment of biomass burning optical properties).

Experimental

12865 (20) : vegetation should not by hyphenated.

12865 (19-21) : Can some measure of distance to fire activity be given here? Is the site surrounded by the fire activity or at the margin?

12865 (24): Should be reported as the particle aerodynamic diameter here and elsewhere in the manuscript unless a correction for density has been applied.

ACPD

10, C4865–C4872, 2010

12866 (20) : The potential for negative artifacts (e.g., underestimation of carbonaceous particle mass due to volatilization from filters) should also be mentioned here. There should also be some mention of how these artifacts could affect interpretation of the size-resolved data (e.g., redistribution of semi-volatile organic species across different filter/impactor stages) here or in section 5.1.

12866 (24) : UPR not defined

12866 (28) : Please provide a reference for the aethalometer (e.g., Hansen et al., Sci. Tot. Environ., 1982).

12868 (2) : Were any attempts made to correct the aethalometer observations for measurement artifacts (e.g., Weingartner et al., 2003; Arnot et al., 2005)? How were wavelength differences between the MAAP and aethalometer accounted for (e.g., 1/lambda or some measured value)? The wavelength(s) of both instruments should be provided, particularly the wavelength used to convert the optical absorption to BCe that corresponds to the absorption cross section (should be the specific absorption cross section) of 10 m2 g-1.

12870 (25) : The aethalometer and LTM measurements use different constant to convert the measured attenuation to BCe (10 vs 19 m2 g-1). Is there a physical justification for this? If not the same factor should be used for the conversion.

12871 (20): Please state which filters were these measured on and the diameter range of the measurement. The uncertainty value provided should be defined (e.g., standard deviation, standard error, inter-quartile range, etc.).

12872 (5) : A minor point, but it is probably more accurate to say that the TC observations in the current study were within the range of values observed in the previous campaign rather than being "consistent" with them. How do the averages and medians compare?

12872 (8-11) : Do the individual samples agree well? What was the strength of corre-

ACPD
10, C4865–C4872, 2010

Interactive Comment

lation between the sampling methods?

12872 (25) : Also provide standard deviation or other measure of variability about the mean.

12873 (12) : Changes in embedded soot morphology may also play a role in altering the optical properties of material in the filter.

12874 (3) : In addition to LTM measurements a more direct comparison to the ECw572Âň values would be to use the ATN measured through the filter after the OC has been evolved.

12874 (11) : The potential role of negative artifacts should be added to this discussion.

12874 (21) : If the positive artifact for the HVDS system was only 5% why are the impactor artifacts 3-6 times larger? Can this be attributed to differences in face velocity or other phenomena?

12876 (12) : This is an opportunity to include the IC results in the analysis. . .a strong correlation between EC and potassium would also support this conclusion.

12876 (24) : 5-10% BC by mass.

12876 (29) : Avoid use of the term soot as its definition is somewhat arbitrary, as pointed out in the introduction.

12877 (1) : If available, measurements of excess carbon monoxide and carbon dioxide mixing ratios would support the claims made regarding the role of fire behavior. A comparison to similar measurements for well-characterized fires (e.g., aircraft or laboratory studies) would be helpful. The addition of secondary organic aerosol would lead to lower EC/TC ratios and mask the nature of the fires responsible for the EC emissions and should be mentioned.

12877 : Is this the same method as presented by Hadley et al. (Environ. Sci. Technol., 2008)? If so reference should be given. It is not clear if BC concentrations were

ACPD

10, C4865–C4872, 2010

Interactive Comment

measured using the method of Hadley et al. (2009) or the earlier single-wavelength approaches.

Section 4 could be combined with the other results. Sections of it are also repetitive and should be removed.

12878 (19) : Please state which time period and/or samples these values represent.

12878 (21) : The OC to POM conversion factor could be compared with the value obtained from a mass closure that compares the measured gravimetric PM2.5 mass to mass reconstructed from measured species.

Section 5 could also be moved to the results section

12879 (9-23) : Move this discussion to the experimental section. Please give an estimate of the magnitude of the size shift.

12880 (15) : Another possibility is a small contribution by gas-phase adsorbed species to the absorption signal.

12880 (20) : Do the IC results confirm this? Not sure the reasoning here is valid. Mixing of BC with SOA etc. will produce the exact same result as the sizing is done on an internal mixture.

12880 (26-28) : I believe the Reid et al. (2005) paper discusses total aerosol volume/mass distributions as opposed to OC and EC mass.

12881 (1) : The comparisons to the droplet mode should include the caveat that the measured particle diameters reported here are aerodynamic diameters. It is also difficult to compare the results to previous studies without a discussion of the uncertainties in diameter due to the stage uncertainty.

12881 (3-16) : Figure 10 support the conclusions presented here and they should be removed. While POM is higher at night for stages between 0.39-2.4 um, it is lower at night for all stages below 0.39 um, nor is BCe “almost the same” for day and night

ACPD

10, C4865–C4872, 2010

stages. Nighttime BCe is at factor of 2 higher than daytime BCe for stages 0.62 um and larger. Though it is difficult to integrate the data by eye it appears that total BC is also higher at night by at least a similar relative magnitude as observed for POM. No uncertainties are given, either, so the significance of the relationship cannot be determined. Do the bulk samples support this conclusion? What about the average of all day and night size-segregated samples?

12881 (16) : Additional discussion is needed to assess the role of water uptake on the sizing results. What was the relative humidity during sampling. Can some estimate of hygroscopic growth be made based on previous measured smoke measurements and used to adjust the measurements to "dry" diameters?

12882 (17) : This conclusion is not supported by the evidence provided in the paper.

Tables: General comment – the number of significant figures reported is different for

different methods. . .are these techniques really more precise than those reported with fewer significant figures?

Table 1 : The aethalometer results should be shown next to the fine or coarse mode integrated values rather than on the same row as the first stage.

Table 2 : STDEV should be written out or abbreviated differently.

Table 3 : Please also report the standard deviations.

Figures

Figures 1 and 9 should be combined. The x- and y-range of Figure 1 is larger than necessary. Please provide error bars for each stage if possible.

Figure 2: Change "more significant" to "relatively larger" or similar in the caption.

Figure 4: Provide error bars by propagating the uncertainty in TC measured by each system. Bottom labels of the figure are being chopped off.

ACPD

10, C4865–C4872, 2010

Figure 6: remove shadows from the plotting symbols. Different symbols should be used in 6a to give the different EC measurement methods.

Figure 7 is unnecessary and results could be easily described in the text.

Figure 9: Better to show OC rather than POM because POM requires the assumption of a constant OC to POM ratio for all sizes.

Interactive comment on Atmos. Chem. Phys. Discuss., 10, 12859, 2010.

ACPD

10, C4865–C4872, 2010

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Discussion Paper