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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Long-term Brown Carbon and Smoke Tracer Observations in Bogotá, Colombia: Association to Medium-Range Transport of Biomass Burning Plumes" *by* Juan Manuel Rincón-Riveros et al.

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Anonymous Referee #2

RC: This paper investigates the contribution of biomass burning from distant locations to air quality in Bogota Colombia based on an extensive data set of aerosol light absorption at multiple wavelengths. Most data reported are from a measurement site upwind and at higher elevation than the city. Filter measurements of smoke tracers are also used to support the analysis, along with satellite-based fire counts and air mass

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Discussion paper

back trajectories. Overall the paper is a nice contribution to an understudied location and appropriate for publication in this journal. The results are interesting and the analysis very thorough, however, some components are confusing and should be clarified. I agree with the other two reviewers that the sensitivity of the reported results to the choice of AAE for BC (AAE=1) and for BrC (AAE=2) should be assessed. A value of BrC AAE of 2 seems especially arbitrary. It is not clear to me why the authors utilized this analysis method at all since it adds unnecessary complexity and ambiguity; more related to this question follows below.

Authors Response: This comment is common to all reviewers. We addressed the issue of uncertainty in two ways: 1- By expanding and explaining the uncertainty associated to the attribution of absorption to BB and FF (including sensitivity analysis) and 2- by discussing potential uncertainties associated to transforming babs into eBC and BrC. Figures were modified to include an axis with babs (in Mm-1) We also expanded the supplementary material to clarify and make more transparent how the separation between BB and FF was performed.

RC: Why was 470 and 880 nm light absorption data used in the fractional biomass burning calculation? Explicitly state the reason. e.g, why not 370 and 950 nm, respectively? Similarly, why was 880 nm used for eBC, not 950 nm? Why not use all the wavelength data in some way, instead of just selecting a few wavelengths from the measurements (more on this below)?

Authors Response: We did not use data from the 370 nm channel since its noise to signal ratio is higher than that of other channels. To analyze BrC/BC typically a short wavelength and a near-IR wavelength are used. The choice of 880 nm for BC was done as this is what has been historically reported in previous aethalometer studies. BC at 880 and 950 nm channels is almost identical for our data. This is now explicitly mentioned in the manuscript: “The Angstrom exponent was computed using a wavelength in the near-UV, where absorption from some organic compounds can be significant, and a near IR wavelength, where absorption is dominated by black carbon. However,

as the 370 nm channel had a larger noise to signal ratio, the limit of detection of this channel was considerably higher and was not used in the analysis. Equation 1 was then applied to babs measured at 470 nm and 880 nm wavelengths to compute an observed α."

We also performed some sensitivity analysis on the pair of channels used to calculate the Angstrom exponent and evaluated its impact on our results. The mean Angstrom exponent varies slightly according to the wavelength pair chosen (α_(450nm-950nm)=1.025$\pm$0.2 and α_(450nm-880nm)=1.065$\pm$0.22) affecting the absolute value of inferred f_BB and BrC. However, because most of our analyses are based on correlations and associations to fires, these remain unchanged.

RC: Why does one even need to calculate a BC and BrC concentration, instead of just using the absorption coefficient? For example, simply using the absorption measured at a high wavelength as a tracer for BC and Abs measured at a low wavelength (e.g., 370, or if too noisy, 470 nm) as a tracer for BrC, after the Abs by BC at that wavelength is removed. This can be done by assuming a BC AAE of some value, such as 1. This seems like a much more transparent way to apportion BC and BrC from the multiwavelength Aeth data and it eliminates the need to assume a characteristic BrC AAE. It also simplifies an uncertainty analysis on the sensitivity of the results to only the value of BC AAE. It would be interesting to see a correlation between the BrC mass inferred by the method in this paper and the BrC abs at some wavelength (eg, 370 nm).

Authors Response: We are aware of the challenges of performing this decomposition. The method proposed by the reviewer is plausible, but it disregards contributions to absorption from BrC even at high wavelengths. The method we employed accounts for the contribution of BrC and BC absorption at all wavelengths. We now discuss this much more thoroughly in the manuscript. To address the valid concern regarding calculations of BC and BrC, we now added babs (Mm-1) in a secondary axis in Figure 2b and 2c. Since eBC and BrC are both proportional to babs, their correlation to the other datasets remains unchanged.

RC: Instead of picking a specific wavelength for BrC why not use all the Abs vs wavelength data. That is, fit the data with an AAE using all the wavelength and then use the fit to predict BrC AAE (data AAE-1) and then determine light absorption at some low wavelength with fit AAE-1.

Authors Response: During the data analysis phase, we did explore a variety of multi-wavelength methods. However, most of those methods require and even greater number of assumed parameters (e.g., Massabó et al., 2015). In their approach 5 wavelengths are used, but there is the need to assume three parameters and solve for three more. We tested this method and when applied to our data was much more sensitive to parameters choice than the simpler method we employed.

RC: Light absorption data are based on PM1, chemical composition and mass on PM2.5. PM1 was chosen to reduce possible influence of dust light absorption on the inferred BrC mass. The authors could test if there is any correlation between dust (eg, Ca2+) and BrC.

Authors Response: This is an excellent suggestion. However, we do not have Ca2+ data available in our samples. We think there is strong evidence showing that our BrC observations are strongly linearly correlated with levoglucosan and other BB tracers, suggesting its origin is indeed from biomass burning.

RC: Line 236-237: This line is unclear, suddenly there is a discussion that changes from eBC to EC. How does this data prove eBC is EC. Why not just say that eBC is from urban traffic and industrial emissions? Also, why is EC only assumed to be from these two sources?

Authors Response: Corrected. The phrase now reads “The strong correlation between both datasets suggests that eBC at the Monserrate site is closely associated to urban emissions. According to a recent emission inventory in Bogotá, mobile and industrial emissions are the dominant primary particle sources in the city. Furthermore, cargo and public transportation have the largest emissions share, and most of those vehicles

are diesel powered (Pachón et al., 2018)."

RC: Line 248-249, first line after heading 3.1. This line is unclear. Is the eBC, BrC and fire counts data (Fig 3b) from the hill top site and the PM2.5 mass (Fig 3a) from the urban air quality stations in the city? That means that Fig 3a has data from two different sites? This complicates the comparison and the discussion that follows this line. More clarity is needed here. Please specify on the plots in Fig 3 what site the data is from.

Authors Response: This is correct. We tried to be as clear as possible in the caption and throughout the text. Now Figure 2 in the manuscript has been modified to include the origin of the data (i.e., panels (b) and (c) are marked Monserrate site). Similar changes were performed on Figure 3. However, we want to emphasize to the reviewer that the aim of Figures 2 and 3 in the paper is to show that our BC measurements are strongly correlated to PM2.5 in the city, while BrC measurements do not. Instead, BrC resemble regional fire counts according to their correlation coefficients.

RC: Line 282, typo change that to local emissions, to, than to local emissions.

Authors Response: Corrected.

RC: Line 289-290 states, ... However, optical methods are not always quantitative methods to determine BB aerosol loading. What is this statement based on?

Authors Response: The statement originally meant to refer to the issue of translating absorption coefficient data into concentrations (which, as the reviewer pointed out, require the assumption of a MAC). That is what we originally meant by "quantitative". This whole paragraph is now rewritten and now reads: "However, due to the uncertainties in mass absorption cross sections, aerosol absorption measurements are not always straightforward to translate into BB aerosol concentrations. To establish the relationship between our Aethalometer based BrC (Section 2.2) and analytical methods to quantify BB aerosols (Section 2.3), we compared. . .."

RC: Line 314. Is this true; the Monserrate site (also called at times, the hill top site)

maybe a fairly close distance to the urban center, but it is decoupled from the city at times due to its higher elevation and changes in BL height. This mixing of the hill top site with the urban site throughout the paper leads to confusion. Often the term monitoring site is also use, which is apparently the Monserrate site, not the urban air quality sites? I suggest being more specific and consistent throughout the paper on what the sites are called.

Authors Response: Point is well taken. Our monitoring site is now referred to as Monserrate Site consistently throughout the whole manuscript. We also included a paragraph in the Methods to make explicit that our data comes from two different sources: our station at Monserrate (for eBC, BrC, and smoke tracers), and the AQ monitoring sites in the city (for PM2.5 only).

RC: Last line of Conclusions. What is the 13% based on, mass ratio of eBC and BrC. This is then not an optical ratio and should be noted, it may also depend on how BrC was determined (AAE=2). Again, calculating mass concentrations of BC and BrC from the absorption data just leads to confusion and more uncertainty, in my view.

Authors Response: The 13% is based on f_BB. With the expanded and improved Methods and Supplementary materials we show that f_BB=(b_(abs,BB) (λ))/(b_abs (λ)), i.e., the ratio of b_(abs,BB) (λ) We performed a sensitivity analysis of this monthly mean f_BB with α_BB=2.0$\pm$0.4 and α_FF=1.0$\pm$0.1. We know report a range in this percentage. It now reads: “During our observation period, the month with the largest contribution of BB aerosols to light-absorbing material was March with 10%$\pm$5%. The largest contribution was identified for February and March 2019, with 13%$\pm$6%. The uncertainty estimates in this fraction are due to uncertainty on the assumed absorption Angstrom exponent for biomass burning and fossil fuel burning used in the attribution algorithm”

Please also note the supplement to this comment:
https://www.atmos-chem-phys-discuss.net/acp-2019-1124/acp-2019-1124-AC3-

supplement.pdf

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