Spectral absorption of brown carbon in the Central Amazonia

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Abstract

There is a need to better characterize aerosol absorption due to its remarkable radiative effects on climate. In particular, we need to understand the separation of total absorption between the two main components, the black carbon (BC) and brown carbon (BrC), especially in places where the anthropic influence is little, such as in pristine regions like Central Amazonia. The mechanisms that control the formation and evolution of BC and BrC in tropical forests remain unclear. In this study, we have performed detailed measurements at the Amazon Tall Tower Observatory (ATTO) tower on aerosols collected in Nuclepore filters and analyzed them with high-resolution optical spectrometers with a wide spectral range (300 to 2500 nm). Thus, we determined the absorption characteristics of BrC as a function of wavelength. The results show that BrC absorption is spectrally significant below 660 nm and is maximum at wavelengths close to 370 nm. Combining the measured spectral dependency with MIE modeling of the BC contribution, we determined that the BrC accounts for 14.8% of the total absorption. A similar fraction of BrC to total absorption was obtained through a similar analysis of in-situ measurements. Elemental chemical analysis of the filters, cluster, and factor analysis shows that the BrC is associated with airborne dust. The different methods to quantify BC and BrC are consistent and show similar results. This study will allow the quantification of the role of BrC and BC in aerosol absorption of radiation in Amazonia.

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ABSTRACT

There is a need to better characterize aerosol absorption due to its remarkable radiative effects on climate. In particular, we need to understand the separation of total absorption between the two main components, the black carbon (BC) and brown carbon (BrC), especially in places where the anthropic influence is little, such as in pristine regions like Central Amazonia. The mechanisms that control the formation and evolution of BC and BrC in tropical forests remain unclear. In this study, we have performed detailed measurements at the Amazon Tall Tower Observatory (ATTO) tower on aerosols collected in Nuclepore filters and analyzed them with high-resolution optical spectrometers with a wide spectral range (300 to 2500 nm). Thus, we determined the absorption characteristics of BrC as a function of wavelength. The results show that BrC absorption is spectrally significant below 660 nm and is maximum at wavelengths close to 370 nm. Combining the measured spectral dependency with MIE modeling of the BC contribution, we determined that the BrC accounts for 14.8% of the total absorption. A similar fraction of BrC to total absorption was obtained through a similar analysis of in-situ measurements.

Elemental chemical analysis of the filters, cluster, and factor analysis shows that the BrC is associated with airborne dust. The different methods to quantify BC and BrC are consistent and show similar results. This study will allow the quantification of the role of BrC and BC in aerosol absorption of radiation in Amazonia.

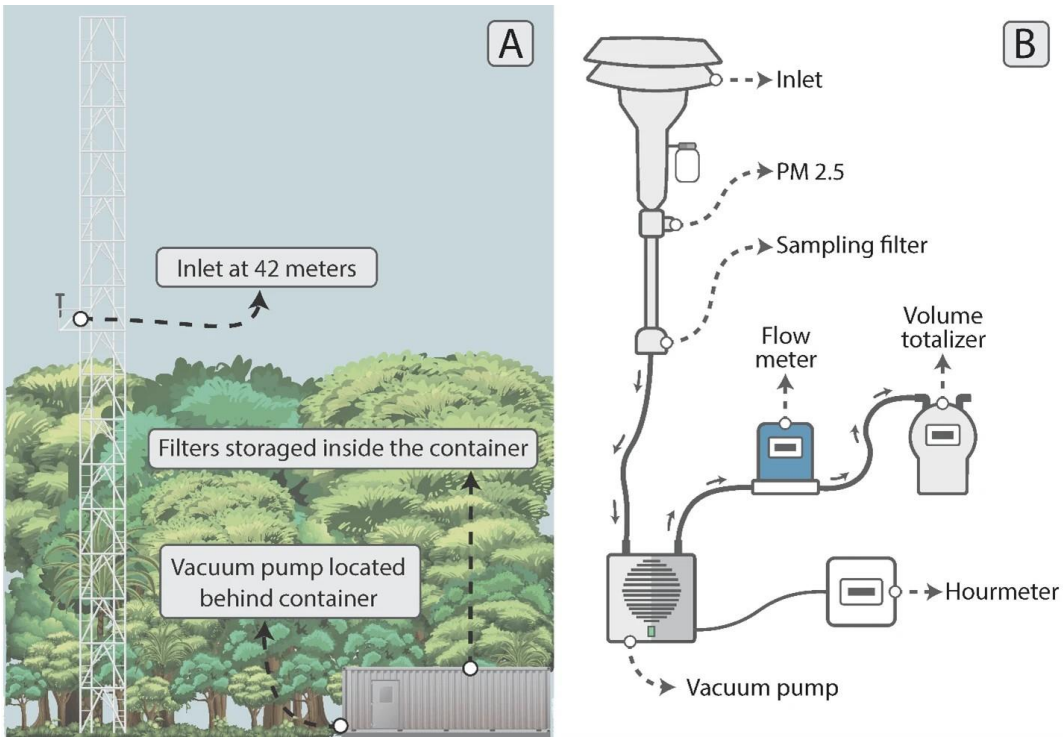
SUMMARY

The mechanisms that control the formation, evolution, and long-range transport of BC and BrC in tropical forests remain unclear. In this study, we have performed detailed measurements at the Amazon Tall Tower Observatory (ATTO) on aerosols collected in Nuclepore filters and analyzed them with high-resolution optical spectrometers with a wide spectral range (300 to 2500 nm). Thus, we determined the absorption characteristics of BrC as a function of wavelength.

METHODOLOGY

An inlet sampler with an aerodynamic cut in PM2.5 was used, with a flow of 17 LPM, installed at a height of 42 meters in the Instant Tower, located at the ATTO site, with coordinates 2°08.6470 S and 58° 59.9920 W (WGS 84). Each sampling was done over approximately 7 days, varying with weather conditions and sampled mass. A total of 34 filters were collected between March 26 and October 31, 2019, where we obtained samples from the dry period with a strong influence of burning, wet season rainy period, and also episodes with long-range transported dust from Sahara to Central Amazonia.

Figure 1: a) Graphic representation of the sampling of the filter on the ATTO website, b) detailed diagram of the sampling of Nuclepore filters.



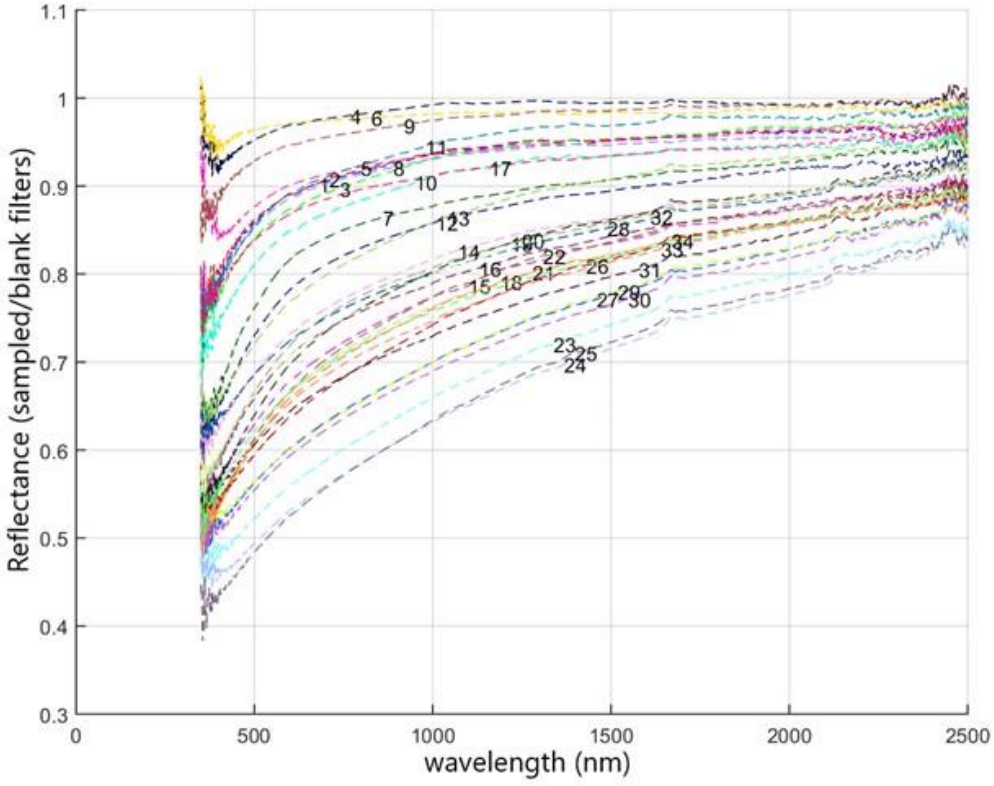
The spectral light absorption of the sampled collected filters was investigated by

measuring the reflectance of the Nuclepore filters comparatively to blank filters reflectivity. Two spectrometers with different broadband illuminators were used: an Avantes AvaSpec 2048, with ultra-violet (UV) starting from 300 nm to near-infrared wavelengths (NIR) up to 1100 nm, with a high-power UV-VIS light source from Hamamatsu model L10290, and a FieldSpec Pro from Analytical Spectral Devices with a range of 350 to 2500 nm with a reflectance lamp from ASD Inc.

RESULTS

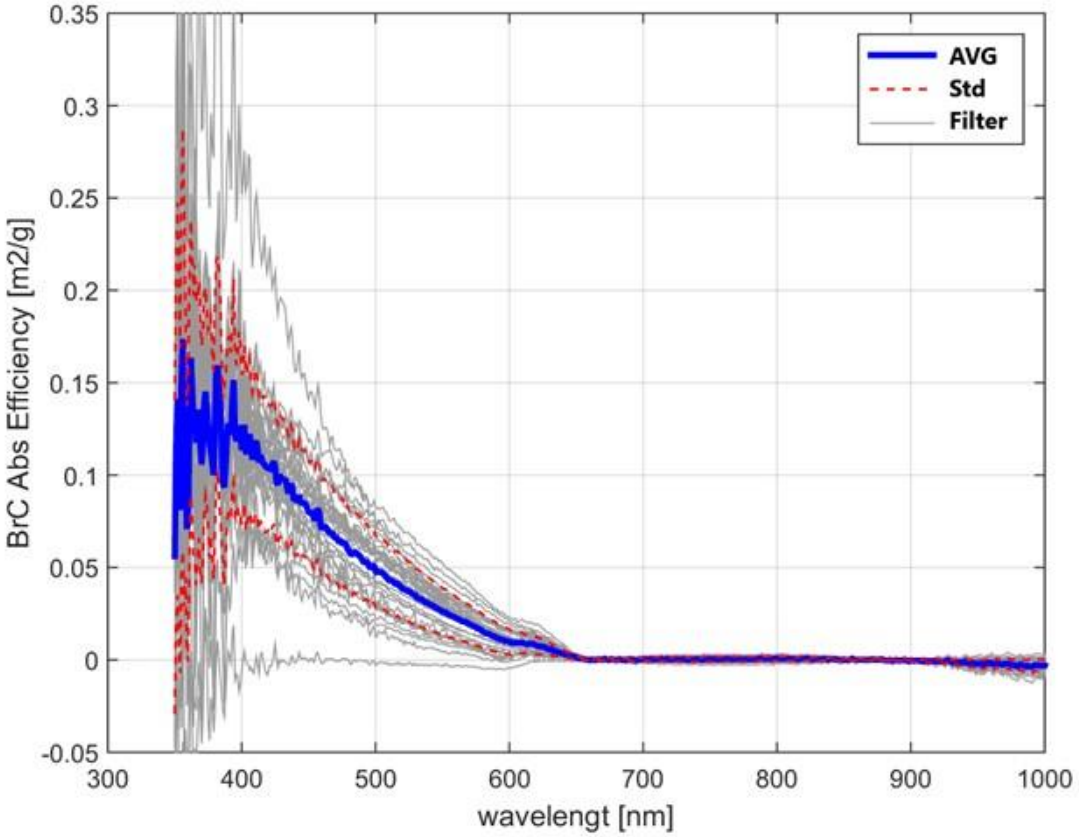
From the high-resolution optical spectrometer data, we obtained the reflectance curves of the aerosol-loaded filters. We analyzed also the blank filters, to subtract their effects.

Figure 2 Reflectance of 34 filters sampled at Amazonia ATTO site obtained by the two high-resolution optical spectrometers from 350 – 2500 nm.



Using the method of Martins et al., 2009, it was possible to calculate the spectral absorption efficiency in m^2/g of the deposited aerosol. We then separate the BC and BrC portions using the method developed by Wang et al., 2016. This procedure allowed us to obtain the BrC spectral variability behavior for each filter.

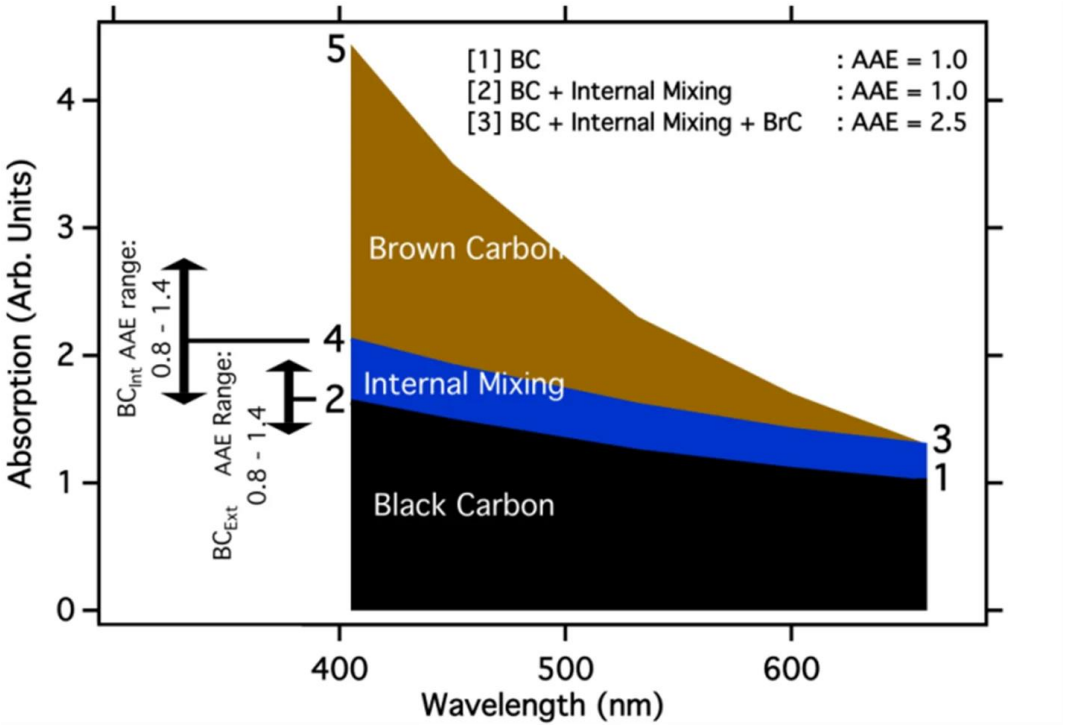
Figure 3 Absorption efficiency of BrC calculated for each of the 34 filters sampled in Amazonia at the ATTO tower during March-October 2019.



DISCUSSION

A study by Lack; Langridge, 2013, used Mie code showing that the contribution of BrC grows with the decrease in wavelength, and may even be the largest absorption component for lambda close to 400-450 nm. BC's contribution, on the other hand, is fairly constant as a function of wavelength.

Figure 4 Illustration of absorption contribution as a function of wavelength, showing how different Ångström Absorption Exponent (AAE) is used to characterize the different components. Brown Carbon dominates absorption for near UV wavelengths.



Our study shows through real measurements at sampled filters at the ATTO site in central Amazonia that the BrC fraction of absorption varies as a function of the wavelength (figure 3). The BrC/BC ratios vary significantly for the wet and dry season, as well as variability also caused by different local or long-range transported components dominating the aerosol population.

CONCLUSIONS

Using a method for obtaining and analyzing the spectral variability of aerosol absorption, it was possible to observe a variety of atmospherically relevant BC and BrC properties. In particular, it was possible to observe that assuming a wavelength-independent refractive index and optical properties value does not explain the spectral dependence of absorption. Analyzes with in situ measurements in the ATTO tower with Nuclepore, $PM_{2.5}$ filters in a wide wavelength range (300 to 2500 nm) with high-resolution optical spectrometers allowed us to understand better the behavior of BrC as a function of wavelength.

In our method, we assume that the BrC absorption contribution is negligible from wavelengths of 660 to 2,500 nm. This is supported by laboratory measurements (Chen and Bond, 2010; Zhang et al., 2013; Yang et al., 2009; Kirchstetter et al., 2004).

The average fraction of BrC to BC at 470 nm for the ASD optical spectrometer was 15.9%. The same analysis for the AVA spectrometer was 17.5%. This means that BrC absorption is an important component in Central Amazonian aerosols.

With independent measurements using AERONET sunphotometers, it is also clear the relevance of BrC absorption in Amazonia, where it can reach values as high as 25% of total aerosol absorption. This is a significantly higher percentage, that could be attributed to the fact that AERONET measures the whole aerosol column, while our measurements were at the surface.

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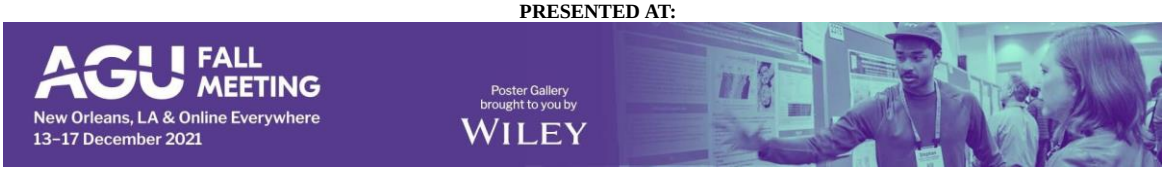

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