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Long-term measurements of aerosol optical properties and radiative forcing (2011-2017) over Central Amazonia

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**Highlights**

- The columnar and surface aerosol optical properties were combined and analyzed.
- A ten-fold increase in the scattering coefficient is observed in the dry season.
- AERONET SSA values were higher 8.4 ± 2.9% compared to the SSA from situ measurements.
- A SWARF AERONET-SBDART regression analysis shows a high correlation.
- SWARF$_{TOA}$ is observed to be -3.66 ± 1.59 W.m$^{-2}$ in the wet season.

**Graphical Abstract**
Abstract

The Amazon region is one of the most pristine continental areas whose concentrations of atmospheric trace gases and aerosol particles are very low, mainly in the wet season. This study provides observational results of aerosol optical and radiative characteristics in-situ as well as atmospheric columnar at a pristine forest in Central Amazonia. Spectral variation of the aerosol properties (Aerosol Optical Depth - AOD), Single Scattering Albedo - SSA, and Asymmetry Parameter - AP) were evaluated using the AErosol RObotic NETwork (AERONET) data. The SSA values under natural atmospheric column conditions (AERONET) were compared to the SSA values calculated with in-situ measurements. The values of Shortwave Aerosol Radiative Forcing (SWARF) on Top of the Atmosphere (TOA) and the SURface (SUR) were estimated using the SBDART model and were validated with the AERONET values with regression analysis. SWARF had a high correlation to TOA (0.97) and SUR (0.92), including dry and wet seasons. Monthly, seasonal and annual mean values of SWARF$_{TOA}$ and SWARF$_{SUR}$ were negative while SWARF$_{ATM}$ values were positive. SWARF$_{TOA}$ was $-9.18 \pm 2.80 \ \text{W.m}^{-2}$ and SWARF$_{SUR}$ was $-20.77 \pm 5.04 \ \text{W.m}^{-2}$ in the dry season, inducing a Heating Rate (HR) of $0.37 \pm 0.13 \ \text{K.day}^{-1}$. This study showed that, for a long series of measurements, the effects caused by aerosols on the radiative flux in the pristine forest of Central Amazonia were of the order of SWARF$_{TOA}$ of $-3.66 \pm 1.59 \ \text{W.m}^{-2}$ and SWARF$_{SUR}$ of $-11.86 \pm 2.35 \ \text{W.m}^{-2}$ during the wet season.

Keywords: 1) Single scattering albedo, 2) Aerosol radiative forcing, 3) Rainforest, 4) Heating rate.

1. Introduction

The impacts caused by atmospheric aerosols on climate change have received special attention during the last decades (IPCC, 2013). Aerosols affect the radiation budget, altering Earth’s climate by absorbing and scattering radiation (direct radiative effect), acting as clouds condensation nuclei, and modifying their physical properties (indirect radiative effect) (Ramanathan et al., 2001; Kaufman et al., 2005; Scott et al., 2018a). The aerosol effects (direct and indirect) have large regional variations, mainly due to short lifetime, complex chemical composition, and interaction in the atmosphere, resulting in high space-temporal heterogeneities (Rajeev and Ramanathan, 2001). Different regions, characterized by different aerosol sources as well as different meteorological conditions can provide important
information on the radiative effects of aerosols at a regional and global scale (Artaxo et al., 2013; Hooda et al., 2016; Bibi et al., 2017; Martin et al., 2016).

The terrestrial biosphere is an important source of natural aerosols (Hallquist et al., 2009; Carslaw et al., 2010; Scott et al., 2018a, b). These natural sources (biogenic volatile organic compounds) can dominate ambient aerosol in the tropics (Pöschl et al., 2010; Martin et al., 2010; 2016). Because natural aerosol constitutes a large fraction of ambient aerosol, it can have important radiative effects (Rap et al., 2013; Rizzo et al., 2013; Scott et al., 2014). The Amazon basin is a great “laboratory” for studying atmospheric processes characteristic of natural conditions, as they existed before the impact of industrialization in the regional and global atmosphere (Andreae, 2007; Artaxo et al., 2013; Martin et al., 2016; Rizzo et al., 2018). The Amazon region is a key ecosystem to understand the climate system but has forest area been decreased by deforestation which began to reach substantial levels in the mid-1970s and continues incessantly until the present day (Andreae et al., 2002; Schafer et al., 2008; Scott et al., 2018b).

The concentration of aerosol in the Amazon is about 300-500 particles per cm$^3$ (Martin et al., 2010), levels that represent an upper limit to the natural atmospheric particle loading before anthropogenic influence (Andreae, 2007; Artaxo et al., 2013; Rizzo et al., 2018). However, the most preserved forest in the region is seasonally influenced by anthropogenic emissions of the biomass burning due to expansion of agriculture, logging, and urbanization (Davidson et al., 2012), also by long-range transport, with contributions from particles outside the basin (Andreae et al., 2015). Thus, the characterization of the optical and radiative aerosol properties of the Amazon in clean and polluted conditions is crucial for our understanding of the dynamic biosphere-atmosphere processes and the resilience of the ecosystem to the changes (Artaxo et al., 2013).

Aerosol Radiative Forcing (ARF) is the net change in the energy balance of the Earth's system due to some forced disturbance in any atmosphere layer (Ramanathan et al., 2001; Singh et al., 2016). Impacts on the radiation balance vary according to the region, so, a comprehensive examination of the optical and radiative properties of aerosols at a local and regional level is therefore necessary (Rajeev and Ramanathan, 2001; Kaufman et al., 2002; Prasad et al., 2007; Bibi et al., 2017). ARF is influenced by the Aerosol Optical Depth (AOD), Single Scattering Albedo (SSA), and Asymmetry Parameter (AP) (Sena et al., 2013). The SSA is the most important parameter that affects ARF and is extremely sensitive to the processes of absorption
and scattering of the aerosols in the atmosphere (Bibi et al., 2017). Small errors in the estimation of SSA can affect the ARF signal (Takemura et al., 2002). The intensity and sign of ARF are also calculated by different types of aerosols, chemical uniqueness, and their size distribution. Therefore, an accurate assessment of ARF is a difficult task and remains more uncertain for the climate system (IPCC, 2013; Bibi et al., 2017).

Several studies in the Amazon basin have already addressed the evaluation of the optical and radiative properties of aerosols (Procopio et al., 2003; Procopio et al., 2004; Patadia et al., 2008; Rizzo et al., 2011; 2013; Artaxo et al., 2013; Sena et al., 2013; Sena and Artaxo., 2015). These studies performed for different periods and used different methodologies to quantify and characterize the disturbances caused by aerosols (Procopio et al., 2004; Patadia et al., 2008; Sena et al., 2013). However, local studies on ARF time series help in an attempt to minimize the uncertainties associated with their estimation. In this study, the aerosol optical and radiative characteristics over the Amazon rainforest were investigated for seven years (2011-2017). The spectral variation of the variables AOD, SSA, and AP measured using AErosol ROboticNETwork (AERONET) data was analyzed. Thereafter, the difference between the SSA data retrieved from AERONET and the SSA calculated using measurements on the ground surface was shown. AERONET radiative forcing products were also used. The monthly, seasonal, and annual variations of Shortwave Aerosol Radiative Forcing (SWARF) on the Top of Atmosphere (TOA), SURface (SUR), and within the ATMosphere (ATM) along with atmospheric Heating Rate (HR) were calculated using Santa Barbara DISORT Atmospheric Radiative Transfer (SBDART) model. A regression analysis of the AERONET-SBDART forcing was also performed to validate the measurements simulated by the SBDART.

2. Site description, measurement, and methods

2.1 Site, instrumentation, and data

This study was carried out with measurements at two sites of the GoAmazon 2014/5 experiment (Martin et al., 2016). The characterization of the optical properties of the aerosols used as input to the radiative transfer code was carried out based on AERONET measurements at site T0e or EMBRAPA (2.8942 °S, 59.9718 °W) (Empresa Brasileira de Pesquisas Agropecuárias) (Barbosa et al., 2014) located 25 km NNW of Manaus (Fig. 1a), a developing city with a population of 1.8 million people (IBGE, 2015). There is little biomass burning activity in the region, but the site is affected by the regional transport of biomass burning pollutants, especially in the dry season (Rizzo et al., 2013). The values obtained for the Single
Scattering Albedo (SSA) of the atmospheric column at T0e were related to in situ measurements at the site T0a (2.1466 °S, 59.0050 °W) the Amazonian Tall Tower Observatory (ATTO; Andreae et al., 2015), located 150 km to the northeast of Manaus (Fig. 1a).

[Figure 1. a) Location of the study area. The yellow marks are T0a and T0e sites. The red mark is Manaus in the State of Amazonas, Central Amazon, Brazil, b) micrometeorological characterization for the T0a, showing the monthly averages for precipitation (PRP), air temperature (Tair), relative humidity (RU), and shortwave radiation.]

In this study, the dry and wet seasons were defined through the AERONET data AOD time series and aerosol scattering and absorption coefficients at different wavelengths, according to Rizzo et al. (2013). The dry season was defined from September to December, while the wet season was from March to June. This definition may not be strictly correct from a climatological point of view, the beginning of each season changes from year to year, depending on precipitation patterns. However, considering the purposes of the interpretation of the aerosol dataset, this is a reasonable choice (Rizzo et al., 2013; 2018). In this way, it is possible to evaluate more clearly the impacts caused by aerosols from biomass burning (dry season), as well as the effect of natural biogenic aerosols (wet season). Figure 1b shows the micrometeorological characterization for the site T0a, showing the monthly values for precipitation (PRP), air temperature (Tair), relative humidity (RU), and shortwave radiation.

In this paper, the data used were retrieved by CIMEL sun/sky radiometers part of the AERONET global network. Measurement of direct sun radiation was done in eight (340-1640 nm) spectral channels with an uncertainty of ≈ 0.01-0.02 for AOD recoveries (Eck et al., 1999), and the amount of water vapor column is measured at 940 nm (Holben et al., 2001). In addition, inversion products were recovered in four (440, 670, 870, and 1020 nm) spectral channels (Dubovik et al., 2002). Data is available in three levels of quality: level 1.0 contains unpatched data, level 1.5 contains data that is tested in the cloud, and level 2.0 data that is tracked in the cloud and is guaranteed quality and can be downloaded from the AERONET website (http://aeronet.gsfc.nasa.gov/), level 2.0 data undergoes a final calibration process with information specific to each site, which is detailed by Dubovik et al. (2002). In this study, 2.0 level data of AOD, SSA, and AP are inversion products retrieved from a selected sunphotometer over T0e (2011-2017). Besides being used for spectral analysis, they were also used as an input to the SBDART radiative transfer code. Before performing analysis, the AOD values between T0a and T0e (between the years 2016 and 2017) were compared, assuming that the aerosol
concentrations were similar for the two sites. Thus, a long time series (T0e) were considered. Direct products were also used, AOD 500 nm total, coarse and fine fractions, precipitable water, and angstrom exponent 440-870 nm.

Surface measurements were performed at site T0a from 2014 to 2017 with a 5-minute temporal resolution as well as daily and 15-day averages for the analysis of the time series. All measurements were taken under dry conditions (RH 30–40 %), assured by an automatic diffusion dryer (Tuch et al., 2009). Inlet lines ran from the measurement level (80 m a.g.l., about 51 m above the canopy height) to an air-conditioned container at ground level, the inlet particle size cutoff was PM 2.5. The aerosol particle scattering coefficients were measured using a three-wavelength integrating nephelometer (Aurora Ecotech 3000) operating at 450, 525, and 635 nm. Every six months the instrument was calibrated using filtered air and CO₂. Sampling time varied between 1- and 5-min. Data were corrected for truncation errors according to Anderson and Ogren (1998), using the tabulated factors for total scatter as a linear function of Angstrom exponent with no cutoff at the inlet (Rizzo et al., 2011; 2013). Aerosol particle absorption was measured using a MAAP photometer (MultiAngle Absorption Photometer – Thermo Inc., Model 5012) (Petzold et al., 2005). The MAAP reports black carbon (BC) concentrations at 637 nm, which were converted to absorption coefficients assuming a mass absorption coefficient of 6.6 m².g⁻¹, used in the firmware of the instrument. Particle absorption coefficients were measured every minute, and a 5% correction was applied to the data to account for an adjustment of wavelength (Müller et al., 2011).

2.2 Data analysis

The nephelometer scattering measurements (in-situ) were interpolated to the wavelength common to the MAAP at 637 nm. With the paired measurements of scattering coefficients and absorption coefficients, the Single Scattering Albedo (SSA) was calculated at 637 nm using the following equation.

\[
\text{SSA(637)} = \frac{\sigma_s(637)}{\sigma_s(637) + \sigma_a(637)}
\]

(Eq.1)
The SSA values as well as the absorption and scattering coefficients were calculated monthly. In addition, the SSA data calculated using Eq. 1 were compared with the SSA obtained at 675 nm from the AERONET station.

The AERONET optical parameters (AOD, SSA, and AP) were analyzed for seasonal variations. These optical parameters fed the SBDART radiative transfer code. Simulations with the SBDART returned the radiative fluxes to the surface and top of the atmosphere. The SBDART outflows (TOA and SUR) were compared to the fluxes estimated by the AERONET algorithm. Furthermore, SWARF (TOA and SUR) instantaneous values were validated according to the AERONET estimates using the statistical parameters: coefficient of determination ($R^2$, Eq.2), Mean Absolute Error (MAE, Eq. 3), Root Mean Square Error (RMSE, Eq. 4) and Willmott’s coefficient (WILL, Eq. 5):

\[ R^2 = 1 - \frac{\sum_{n=1}^{N} (SWARF_{\text{AERONET} n} - SWARF_{\text{SBDART} n})^2}{\sum_{n=1}^{N} (SWARF_{\text{AERONET} n} - \bar{SWARF})^2} \]  

(Eq.2)

\[ MAE = \frac{1}{N} \sum_{n=1}^{N} |SWARF_{\text{SBDART} n} - SWARF_{\text{AERONET} n}| \]  

(Eq.3)

\[ RMSE = \sqrt{\frac{1}{N} \sum_{n=1}^{N} (SWARF_{\text{SBDART} n} - SWARF_{\text{AERONET} n})^2} \]  

(Eq.4)

\[ WILL = 1 - \frac{\sum_{n=1}^{N} (SWARF_{\text{SBDART} n} - SWARF_{\text{AERONET} n})^2}{\sum_{n=1}^{N} (SWARF_{\text{SBDART} n} - SWARF_{\text{AERONET} n} + |SWARF_{\text{AERONET} n} - SWARF_{\text{AERONET} n}|)^2} \]  

(Eq.5)

In this study, SWARF was defined as the difference between calculated net fluxes with and without an aerosol load in the atmosphere. The average daily SWARF calculations in TOA and SUR were performed using the SBDART model for the study period. Radiation fluxes were used to obtain SWARF_{TOA} and SWARF_{SUR} for up and down at one-hour intervals for a period of 24h with and without aerosol conditions separately. The radiative forcing in the TOA and the surface were obtained as the difference between the downstream and upstream fluxes, with and without aerosols. The averaged SWARF is often expressed as Eq. 6. The fluxes were calculated for the solar spectrum (0.20-4.00 µm) for a vegetated surface. The solar spectrum was chosen because of later comparisons with the AERONET fluxes that are estimated by the GAME (Global Atmospheric Model) code (Dubuisson et al., 1996; Roger et al., 2006).

\[ SWARF = \int_{0}^{24} (\text{Flux with aerosol} - \text{Flux without aerosol}) dh \int_{0}^{24} dh \]  

(Eq.6)
The SWARF\textsubscript{ATM} values were estimated as the difference between the radiative forcing at TOA (SWARF\textsubscript{TOA}) and SUR (SWARF\textsubscript{SUR}) using the following equation:

$$\Delta \text{SWARF}_{\text{ATM}} = \text{SWARF}_{\text{TOA}} - \text{SWARF}_{\text{SUR}}$$  \hspace{1cm} (Eq.7)

The SWARF\textsubscript{ATM} in W.m\textsuperscript{-2} represents the amount of solar radiation trapped in the atmosphere by aerosols. The higher the amount of $\Delta \text{SWARF}_{\text{ATM}}$, the higher the amount of energy retained. In this way, the rate of heating due to the absorption of aerosols is calculated from the first law of thermodynamics and hydrostatic equilibrium suggested by Liou, (2002) described as:

$$\frac{\partial T}{\partial t} = \frac{g}{C_p \Delta P} \Delta \text{SWARF}_{\text{ATM}}$$  \hspace{1cm} (Eq.8)

Where $\frac{\partial T}{\partial t}$ is the HR in K.day\textsuperscript{-1}, $g$ is the acceleration to gravity (9.8 m.s\textsuperscript{-2}), $C_p$ is the specific heat capacity of air at constant pressure (i.e.1006 J.kg\textsuperscript{-1}.K\textsuperscript{-1}) and P is the atmospheric pressure difference between the surface and 3 km, where most of the aerosols are. Therefore, P was taken as 300 hPa (Kaskaoutis et al., 2013; Singh et al., 2016; Bibi et al., 2017).

3. Results and discussion

3.1 Variability of aerosol optical properties

The complete time series of AOD 500 nm is shown in Figure 2a, b, for sites T0a (2016-2017) and T0e (2011-2017). The agreement between the AOD measures of both sites culminated in the adoption of the measurements made in T0e as representatives of a pristine forest region, used in the radiative transfer code (SBDART) for simulations of radiative fluxes. Figure 2b shows the seasonality of the optical properties of the aerosols in the study area. The increase of the aerosol load in the dry season causes the increase of the AOD values, where fires cause changes in optical properties and the concentration of aerosols in the Amazon basin (Sena et al., 2013; Artaxo et al., 2013; Rizzo et al., 2013).

[Figure 2. a) Time series for aerosol optical depth (AOD 500 nm) retrieved by AERONET sun-photometer in Central Amazonia "T0a" ATTO, b) "T0e" EMBRAPA and c) linear regression for AOD 500 nm "T0e" and AOD 500 nm "T0a” instantaneous measurements.]
Figure 2b also showed year-to-year variations in the optical properties of the aerosol particles that occur mainly due to variations in the amount of precipitation and in the amount of the sources of biomass burning (Rizzo et al., 2013). The maximum AOD (1.8) was observed in 2015. However, no maximum AOD reached values higher than 1.0 for other years. This trend was due to an increase in the number of fires in the same period, where there was an increase of approximately 26% (from 84 to 115 number of fires) from 2014 to 2015.

The aerosol parameters obtained from AERONET station depend on the number of fires in the Amazon Forest. These fires release large amounts of particles and gases into the atmosphere which influence the chemical composition, optical properties of aerosols, and influence directly the forcing of the radiative fluxes. To complement the analysis of variations in the optical properties of aerosols, Figure 3 shows the average 15- days of the parameters measured by AERONET and measures in-situ, as well as the monthly distribution of fires over the legal Amazon and the states of Amazonas and Pará.

Additionally, the seasonal variation observed for the optical properties of aerosols in the atmospheric column can also be seen on the surface. Figure 3 shows that the particle scattering coefficients increased substantially under the influence of biomass burning particles. This is a consequence of the increased fine-particle concentration in the dry season, which is more efficient at scattering light compared to the coarse mode dominated biogenic particles in the wet season (Figure 3b). There is an observed difference between the scattering coefficients values for the years 2014 and 2015 in Figure 3, the scattering coefficients reached maximum values between 160 and 180 Mm$^{-1}$ in 2014, while the maximum value in 2015 was 600 Mm$^{-1}$. The increase in scattering coefficient values from 2014 to 2015 was already expected due to the substantial increase in the biomass burning records that occurred in the Amazon basin during the same period. Although there were no fire outbreaks at the forest site, it was influenced by regional emissions of biomass burning carried by air masses (Rizzo et al., 2011; 2013).

Deforestation fires can smolder for days, producing particles at high emission factor rates with small black carbon content, thereby decreasing the aerosol absorption and increasing...
the aerosol scattering of the original plume (Reid et al., 2005). Evidence shows an increased
tendency in the scattering of light by aged particles due to physical and chemical atmospheric
processes, that act in the increasing of particle sizes (Brito et al. 2014). Conversely, in situ
observations of biomass burning indicate that carbonaceous aerosols concentrations normally
decrease by only 10 to 50% of fresh smoke to the regional haze (Capes et al., 2008), through
dilution with cleaner background air. In this way, it is reasonable that the dispersion of particles
and absorption coefficients vary by different factors, depending on the plume characteristics
and atmospheric conditions (Rizzo et al., 2013).

The barplot for scattering coefficients $\sigma_s(\lambda)$, absorption coefficients $\sigma_a(\lambda)$, and SSA
with the wavelength of 637 nm for the years 2014 and 2017 are presented in figure 4. We
observed an increase of ten-folds ($\sigma_s(\lambda)$) from May to October (from 4 to 40 Mm$^{-1}$). The
variation in the absorption by aerosols was 1 to 3 Mm$^{-1}$ for May and August, respectively. These
results are in accordance with Artaxo et al. (2013) to a nearby area, which is also a pristine
forest (TT34, Artaxo, et al., 2013). In the wet season, there is little burning activity, the
absorption coefficients observed at the time are attributed to biogenic particles (Pöschl et al.,
2010; Rizzo et al., 2013). In the wet season, SSA values reached their maximum at the average
of 0.88, while in the dry season it was at an average of 0.86. Although the magnitude of the
averages between the dry season and wet season is close, it is possible to observe that the month-
to-month variation is much more evident, ranging from 0.92 in May to 0.83 in August.

Rizzo et al. (2013) also analyzed the seasonal pattern of SSA in the forest located in the
central region of the Amazon, finding no difference between medians in the dry and wet
seasons. Furthermore, Rizzo et al. (2013) results (SSA = 0.88, for both seasons) resemble the
results obtained in this study (SSA = 0.88 in the wet season and SSA = 0.86 in the dry season).
Low SSA values (SSA = 0.83) found in the wet season are due to low scattering values (driven
by low aerosol loads) combined with the absorption coefficients (driven by the presence of
light-absorbing biogenic aerosols) (Artaxo et al., 2013).

The monthly variation (Fig. 4) of the SSA values was due to microclimatic conditions
(Fig. 1b), transport, and local contributions of biomass burning. Moreover, high SSA variability
found in February may be associated with the influence of mineral dust and aged particles from
biomass burning from the advection of aerosols from Africa. This contribution affects the absorption coefficients, while the scattering coefficients are not significantly affected (Rizzo et al., 2013). In addition, the highest SSA medians were found due to the organic fraction in April and May, which can spread the radiation efficiently. Besides, the high atmospheric moisture content causes an increase in the diameters of the submicron particles, which consequently cause an increase in the scattering and then an increase in the values of SSA (Fig. 3d). The SSA decreased approximately 10% from May to August, this is because of the biomass burning in the neighboring regions and the high absorption power of black carbon. There was a slight increase in the SSA from August to December due to the blending processes between the burning plumes that reached the study area and the local particles. This causes an increase in the scattering of light caused by organic aerosols from biomass burning.

The seasonal pattern of SSA calculated in situ was consistent with AERONET measurements in the atmospheric column (Fig. 4c). According to Anderson et al. (1999), the values of SSA in natural conditions of high humidity for the atmospheric column should be higher than the dry measurements taken on the surface. In this study, an average difference of $8.4 \pm 2.9\%$ between the SSA calculated in situ and AERONET measurements in the atmospheric column was obtained. There was no significant difference between in situ and the atmospheric column, however, there is an observed strong month-to-month difference.

Besides, the lowest difference obtained between the SSA calculated in situ and AERONET measurements was found in May (3.0%). The maximum difference was 11.8% in August and September. Although the monthly pattern for both measurements was similar, the atmospheric water content increased considerably the SSA. Likewise, the SSA medians in the wet season were observed as high as 0.99 in the atmospheric column. It highlights the dominant effect of scattering on the radiation extinction.

### 3.2 Influence of Fire Activities

For illustration purposes, Figure 5 shows the 24-hour compilation of fire outbreaks (INPE, 2018) with the Hysplit trajectory model for October 4, 2015. This illustration supports the hypothesis that the major contribution of biomass burning emissions comes from neighboring regions. The outbreaks of fire in the state of Pará (Fig. 3.a) directly influence the study area. The work of Palácios et al. (2020) about the study area points out that during the change from wet to dry season, the fraction of biogenic aerosols practically does not change,
however the mixtures OC-EC, characteristics of biomass burning, go from 16% to 28% from wet to dry season.

In general, the results presented in Figures 3 and 4 reflect the complex mixture of aerosol during the period of biomass burning in the region, between August and October. During this period, local and regional influences produce Organic Aerosols from Biomass Burning (BBOA) fresh (less-oxidized) that mix with the aerosol from Africa which is quite aged (more-oxidized). This complex mixture strongly modulates optical properties in the Amazon during the months of the year (Saturno et al., 2018; de Sá et al., 2019). External factors, such as El Niño 2015, and the increase in the number of regional fires, also contributed strongly to the significant increase in the scattering coefficient (4-40 Mm$^{-1}$), absorption (1-3 Mm$^{-1}$) values, and the lowest SSA values (0.92-0.83) observed that year. These results are, respectively, in ascending order, from 24 ± 18 to 48 ± 33 Mm$^{-1}$ and from 3.8 ± 2.8 to 5.3 ± 2.5 Mm$^{-1}$, and in the descending order of 0.96-0.86 presented by Saturno et al. (2018), using data from ATTO and ZF2 (Manaus-AM).

The most part of the time (September-October), these variations are intrinsically linked to the 27-47% increase in BrC concentration due to regional fires in South American (AS), although Africa’s BBOA is the main source of pollution at ATTO during the August (Saturno et al., 2018).

Several other mechanisms have also been postulated to explain the changes observed in optical properties, many of them as a function of the chemical composition of the aerosol (not evaluated in this study). For example, the dominant presence of POA (Primary Organic Aerosol) and Long-Range Transport (LRT) of dust from North Africa (Saleh et al., 2103; Pokhrel et al., 2016) may explain the low absorption values observed during the wet season (Fig. 3-4). The presence of POA seems to be more associated with high BC-to-OA (Organic Aerosol) values which are more efficient absorbers of solar radiation in relation to Secondary Organic Aerosol (SOA) (Saleh et al., 2014 de Sá et al., 2019).

A statistically significant relationship between absorption coefficients, BrC, and PM1 (Particulate Matter), along with organic PM1 chemical composition was also found in the Amazon (Brito et al., 2014). Recent studies have shown that light absorption by BrC decreases not only with LRT (Lin et al., 2016) but also due to increases in the O:C ratio (de Sá et al., 2019). In addition, decreases in the concentration of chemical species associated with nitrogen
are more strongly linked to the increase in light absorption. For instance, the percentage contributed by the CxHyOzNC family seems to be higher if compared to the HOA and LO-BBOA factors which are generally associated with important sources of BrC, coming from fresh plumes of urban and BB aerosols (Laskin et al., 2015; de Sá et al., 2019). Compounds containing nitrogen and organic molecules; humic-like substances, nitro-aromatic catechols, and aromatic carboxylic acids are among the main solar radiation absorbing constituents during the biomass burning season in the Amazon (Claeys et al., 2012; Lin et al., 2016; de Sá et al., 2019). This evidence corroborates to the highest absorption values observed in Figs. 3-4, whereas changes in the spreading coefficient are strongly correlated to the oxidation of OA loading during regional transport, and of LRT events from Southern Africa (Brito et al., 2014; Saturno et al., 2018). These patterns are inextricably linked with complex atmospheric processing; photolysis by solar radiation; gas-particle reaction and reactions at the aerosol-cloud interface in the aqueous phase (Saleh et al., 2014; Laskin et al., 2015; de Sá et al., 2019). New studies, however, are still needed to understand the concomitant changes in optical and chemical properties as a function of different types of biomass burning in the Amazon.

3.3 Spectral variation of AOD, SSA, and AP in the atmospheric column

The SWARF estimates require information on the optical properties of aerosols, AOD, SSA, and AP, as well as the variability of their properties along with the spectral range. Particularly, AOD represents the total extinction of the radiation integrated into the atmospheric column, SSA is the ratio between scattered and absorbed radiation fractions, while AP provides information on scattered radiation. These properties are important for the estimation of the radiative forcing of the aerosols and different spectral variations revealing the size of the particles.

The AOD median values, SSA and AP obtained in 440, 675, 870, and 1020 nm were plotted for the dry and wet season from 2011 to 2017 (Fig. 6). Additionally, the spectral variation of AOD is adequate to provide details on particle size distribution, which was also verified using the Angstrom Exponent (AE) shown in Figure 2 (Eck et al., 2010). AOD was strongly dependent on the wavelength during the dry season, decreasing with the wavelength due to the presence of fine particles, whose result was already expected as a consequence of the emissions by biomass burning (Sena et al., 2013; Artaxo et al., 2013). However, the spectral pattern of the AOD was not significant in the wet season, presenting a change in the aerosol
size distribution towards the larger size range, such as coarse particles (Rizzo et al., 2013; Artaxo et al., 2013).

In addition, AOD, SSA, and AP had a smaller spectral dependency in the wet season compared to the dry season (Fig. 6). Low AOD means a low concentration of particles in the atmosphere (Artaxo et al., 2013). There was a small difference in SSA values between dry and wet seasons. Although this difference was not significant, the decrease of SSA in the dry season occurred due to the presence of absorber aerosols emitted by the biomass burning in this period (Rizzo et al., 2011; 2013; Sena et al., 2013).

The decrease of the AOD and SSA with the wavelength in the dry season occurred in different ways. AOD depends directly on the concentration of optically active particles in the atmospheric column (extensive property) while SSA does not depend on the concentration (intensive), but on the absorption and scattering properties of the aerosol. The higher AOD in low wavelengths is due to the increase of the fine particle concentration in this period. Although, biomass burning aerosols are more absorbent in the wet season than natural aerosols. Unlike other regions impacted by high aerosol loads (Kaskaoutis et al., 2012; Singh et al., 2016; Bibi et al., 2017), SSA decays with the wavelength, although the predominance of aerosols is concentrated in coarse particles of aerosols in the wet season, they interact less efficiently with solar radiation (Rizzo et al., 2011).

3.4 Comparison of SWARF (AERONET vs SBDART)

The direct mean radiative forcing of aerosols (SWARF, shortwave aerosol radiative forcing) was evaluated in the pristine forest within the Amazon basin. Seasonal and interannual variability of estimated SWARF through the SBDART with optical parameters of the aerosols provided by AERONET were evaluated. The regressions performed for the instantaneous values of $\text{SWARF}_{\text{SUR}}$ and $\text{SWARF}_{\text{TOA}}$ and the statistical evaluation parameters are shown in Figure 7 and in Table I, respectively. There were instantaneous values below -150 W.m$^{-2}$ for $\text{SWARF}_{\text{SUR}}$ and -70 W.m$^{-2}$ for $\text{SWARF}_{\text{TOA}}$. Regressions were also made for the SWARF (TOA and SUR) values, separating the dry and wet seasons. Regression’s parameters are also shown.
in Table I. The error was slightly higher for the regressions in the dry season. However, in general, the associated errors are relatively lower, and the SWARF estimates had good agreement with the AERONET values.

[Figure 7. Scatter plots of AERONET vs SBDART, SWARF at the a) surface (SUR), and b) on the top of the atmosphere (TOA) in Central Amazonia (T0e) from 2011 to 2017. These were instantaneous measurements.]

For other locations around the globe, recent studies have shown a good correlation between SWARF values comparing AERONET against SBDART. Adesina et al. (2015) had a high correlation in SWARF$_{\text{SUR}}$ (0.95) and SWARF$_{\text{TOA}}$ (0.97) in Gorongosa, Africa. Li et al. (2010) performed the forcing comparisons in TOA and SUR of AERONET and SBDART and had a high correlation (above 0.92) in 25 different stations throughout China during 2006. Alam et al. (2012) reported high correlations of 0.98 and 0.99 compared to Karachi and Lahore in Pakistan, from 2010 to 2011. Kumar et al. (2015) reported a high correlation (0.89 in the SUR and 0.78 in the TOA) between the AERONET and the SBDART SWARF during the monsoon pre-monsoon in Kanpur, India. Similarly, Valenzuela et al. (2012). Recently, Bibi et al. (2017) found moderate correlations (from 0.5 to 0.8) for SUR and TOA in the Indo-Gangetic plains in India.

[Table I. Coefficient of determination, errors (MAE and RMSE), and Willmot’s coefficient between SWARF AERONET vs SWARF SBDART values to SUR and TOA in Central Amazonia (T0e) from 2011 to 2017.]

### 3.4 Seasonal and interannual SWARF 24 h

Atmospheric aerosols are mainly a combination of types of scattering and absorption particles and their impacts in the form of cooling and heating of the atmosphere are a function of different optical properties (Bibi et al., 2017). In this study, SWARF was calculated separately for TOA and SUR using the SBDART model. However, the difference between SWARF$_{\text{TOA}}$ and SWARF$_{\text{SUR}}$ provides the SWARF$_{\text{ATM}}$ representing the amount of energy retained in the atmosphere by the absorption brought about by aerosols and transformed into thermal energy.

The simulation of the SBDART involved the optical properties of aerosols with a one-hour time resolution (2011-2017). The SWARF$_{\text{TOA/SUR}}$ time values were integrated according
to equation (5) resulting in SWARF\textsubscript{TOA/SUR} 24 h. The monthly averages for SWARF\textsubscript{TOA/SUR/ATM} 24 h are shown in Figure 8. It was observed that SWARF\textsubscript{TOA} 24 h monthly averages reached -14 W.m\textsuperscript{-2} and -36 W.m\textsuperscript{-2} for SWARF\textsubscript{SUR} 24 h. However, SWARF\textsubscript{TOA} 24 h in daily values reached values below -30 W.m\textsuperscript{-2} in October 2015 and SWARF\textsubscript{SUR} 24 h below -60 W.m\textsuperscript{-2} in the same period.

[Figure 8. Monthly variation of SWARF 24 h at TOA (green), SUR (blue), and ATM (red) in Central Amazonia (T0e) from 2011 to 2017.]

The seasonal and annual mean for SWARF\textsubscript{TOA/SUR/ATM} are shown in Table II. Their impact caused by aerosols increases significantly in the dry season (Fig. 8). Table II shows the average of the whole period, it also reveals that SWARF\textsubscript{TOA} 24 h practically increases in three times, from -3.66 to -9.18 W.m\textsuperscript{-2} from wet to dry season. This impact is also felt on the surface, but not in the same proportion. SWARF\textsubscript{SUR} 24 h was from -11.86 W.m\textsuperscript{-2} in the wet season to -20.77 W.m\textsuperscript{-2} in the dry season. The variation between years is also shown in Table II. As seen in Figs. 3-4, it was expected that the greatest impacts on the radiative fluxes would occur in the years whose prolongation of the dry season and the largest load of burnt aerosols reached the study area. There were more than 11 thousand fire spots in 2015, which means the largest record since 2009. Only in September of 2015, when more than 5 thousand fires were recorded in the state of Amazonas, which is 90.3% higher than that registered in the same period in 2014 (INPE, 2018). As seen in Figure 8, and observed in Table II, SWARF\textsubscript{TOA} 24 h was -14.06 W.m\textsuperscript{-2} which means approximately five times higher in the dry season of 2015 than the value found in the wet season (-3.95 W.m\textsuperscript{-2}). The consequence of SWARF TOA and SUR 24 h values are the increase in the HR inside the atmosphere, which is also shown in Table II.

[Table II. Seasonal and interannual mean variation of SWARF 24 h at TOA, SUR, and ATM and HR ± Standard deviations in Central Amazonia (T0e) from 2011 to 2017.]

Comparing the results of this studies to other studies, Procopio et al. (2004), Patadia et al. (2008), and Sena et al. (2013) had a spatial approach and they were restricted to a specific period for biomass burning aerosols, while this was a punctual study and it was in the dry and wet season. Considering that the effect of aerosols from biomass burning, is the result of the difference in effects between dry (-9.1 W.m\textsuperscript{-2}) and wet (-3.6 W.m\textsuperscript{-2}) seasons (Tabela II), this study estimates that SWARF\textsubscript{TOA} caused by biomass burning aerosols is -5.5 W.m\textsuperscript{-2}. This value is consistent with Patadia et al. (2008) and Sena et al. (2013). Table III shows the comparison of some studies for SWARF\textsubscript{TOA} due to the effects of biomass burning aerosols.
The monthly averaged values of SWARF$_{TOA}$, SWARF$_{SUR}$, and SWARF$_{ATM}$ were calculated using daily averaged ARF (SWARF 24 h) from 2011 to 2017. Monthly averaged SWARF$_{TOA/SUR/ATM}$ values for the entire study period, as well as HR, are shown in Figure 9. The monthly statistics agree with the seasonal mean values obtained in Table II. Furthermore, the small variations in the monthly averages are observed from July to December. Although the HR seasonal values practically do not vary (Table II), their decrease is clearly observed, with a minimum value in April (Fig. 9c). Variations are evident in the monthly averages. As expected, the HR variation follows the SWARF$_{ATM}$ values. Its decrease is clearly observed, with a minimum value in March and April (wet season).

The radiative and consequently the climatic implications of atmospheric aerosols are analyzed in the form of atmospheric heating rate (HR). As seen in eq. 7, the HR depends directly on the SWARF$_{ATM}$ variation, which depends on the SWARF$_{TOA}$ and SWARF$_{SUR}$ values. It can be seen in Figure 9c that the monthly HR values varied very little, from 0.22 to 0.37 K.day$^{-1}$. However, the mean HR in the dry season of 2015 reached 0.62 K.day$^{-1}$. The mean HR value found for the entire study period was $0.26 \pm 0.08$ K.day$^{-1}$ in the wet season and $0.37 \pm 0.13$ K.day$^{-1}$ in the dry season. They are consistent with values found by Procopio et al. (2004) to the study area. Procopio et al. (2004) (studying two sites of AERONET, Abracos Hill, and Alta Floresta) evaluated that the aerosols of regional biomass burning caused a variation between -5 and -12 W.m$^{-2}$ for SWARF$_{TOA}$ and between -21 and -74 W.m$^{-2}$ for SWARF$_{SUR}$. The difference between surface and atmosphere measures are associated with the absorption by the aerosol smoke layer of 16-62 W.m$^{-2}$, inducing an HR around 0.14-0.53 K.day$^{-1}$.

On a regional scale, the variation of energy retained by the atmosphere has strong implications for changes in temperature and in the thermal structure of the atmosphere. This work addresses a period with few events of high aerosol loading which contributes to a minimal HR variation in the study area, it was found that the biomass burning aerosols SWARF$_{TOA}$ of -9.18 $\pm$ 2.80 W.m$^{-2}$ and SWARF$_{SUR}$ of -20.77 $\pm$ 5.04 W.m$^{-2}$ induces an HR of $0.37 \pm 0.13$ K.day$^{-1}$.

[Figure 9. a) Monthly averages for SWARF 24h (± Standard deviations) and for TOA (green bar), b) SUR (blue bar), c) ATM (red bar), and HR (black line) in Central Amazonia (T0e) from 2011 to 2017.]
4. Conclusion

This study focused on quantifying the impact of aerosols on the shortwave radiation balance in a pristine forest inside the Central Amazon, under cloud-free conditions. In order to understand the radiative impacts of aerosol on the local climate, SWARF in TOA, SUR, and ATM and associated atmospheric HR were calculated using the SBDART model. The analysis was performed using different time scales (monthly, seasonal, and annual) for a long period (2011-2017). The important conclusions of this study are:

1. From the measurements carried out in situ, the scattering and absorption evidenced the seasonal pattern of the aerosol optical properties. Besides, the scattering increased 10-fold from the wet season to the dry season. Absorption values also increased, but not at the same ratio as scattering. SSA values decreased by 10% at the beginning of the dry season (May-August) due to the strong absorption by black carbon and brown at the beginning of the biomass burning period.

2. When comparing the values of SSA in natural conditions of humidity for the atmospheric column (AERONET) with the values of SSA calculated with in situ measurements, it was observed that the AERONET values have a higher average (8.4 ± 2.9 %), with the greatest differences found in August and September (11.8%), this is due to the increased emission of biomass burning in the atmospheric column. In the wet season, SSA medians were found above 0.99, indicating the dominant effect of scattering on the radiation extinction. The water content in the atmospheric column considerably increased the SSA values.

3. Spectral analysis of AOD, SSA, and AP for the atmospheric column showed a smaller spectral dependency in the wet season compared to the dry season. The AOD varied considerably in the dry season, the higher values of angstrom exponent in the dry season indicated that the addition of fine particles of biomass burning contributed to this variation.

4. The regression analysis of AERONET-SBDART at the SUR and TOA revealed that the SWARF values showed a relatively high correlation to the dry and wet seasons. For the whole period of analysis (without separation between dry and wet season) the R² for SWARF<sub>SUR</sub> (AERONET-SBDART) was 0.96 and SWARF<sub>TOA</sub> was 0.89.
5. The average for the season with highest aerosol loads (dry season) SWARF_{TOA} was -9.18 ± 2.80 Wm^{-2}, SWARF_{SUR} was -20.77 ± 5.04 W.m^{-2} and SWARF_{ATM} of 11.59 ± 4.04 inducing an HR of 0.37 ± 0.13 K.day^{-1}.

6. Considering that the impact of aerosols from biomass burning is given by the difference between SWARF in biomass burning conditions and SWARF under natural conditions (free of biomass burning), this study quantifies the net effect of -5.52 W.m^{-2} on the top of the atmosphere for these aerosols, and -3.66 Wm^{-2} for the effect of natural (biogenic) aerosols in the study area.

Acknowledgments

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Table I. Coefficient of determination, errors (MAE and RMSE), and Willmot’s coefficient between SWARF AERONET vs SWARF SBDART values to SUR and TOA in Central Amazonia (T0e) from 2011 to 2017.

<table>
<thead>
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<th></th>
<th></th>
<th></th>
<th>TOA</th>
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<td></td>
<td>$N$</td>
<td>$R^2$</td>
<td>MAE</td>
<td>RMSE</td>
<td>$WILL$</td>
<td>$R^2$</td>
<td>MAE</td>
<td>RMSE</td>
</tr>
<tr>
<td>Wet</td>
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<td>0.94</td>
<td>0.13</td>
<td>1.36</td>
<td>0.99</td>
<td>0.77</td>
<td>1.21</td>
<td>1.41</td>
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<tr>
<td>Dry</td>
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<td>4.80</td>
<td>6.28</td>
<td>0.98</td>
<td>0.78</td>
<td>3.76</td>
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<tr>
<td>All</td>
<td>1045</td>
<td>0.96</td>
<td>3.44</td>
<td>4.49</td>
<td>0.99</td>
<td>0.89</td>
<td>2.58</td>
<td>3.34</td>
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Table II. Seasonal and interannual mean variation of SWARF 24 h at TOA, SUR, and ATM and HR ± Standard deviations in Central Amazonia (T0e) from 2011 to 2017.

<table>
<thead>
<tr>
<th></th>
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<th>SUR</th>
<th>ATM</th>
<th>HR (K day^{-1})</th>
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<td></td>
<td>Wet</td>
<td>Dry</td>
<td>Wet</td>
<td>Dry</td>
</tr>
<tr>
<td>2011</td>
<td>-3.7 ± 0.7</td>
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<td>-10.9 ± 0.5</td>
<td>-16.2 ± 4.5</td>
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<td>2012</td>
<td>-2.5 ± 0.8</td>
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<td>2013</td>
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<td>2014</td>
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<td>-19.7 ± 2.1</td>
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<td>Year</td>
<td>Period</td>
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<td>Dry</td>
<td>24 SWARF &lt;sub&gt;TOA&lt;/sub&gt; (W.m&lt;sup&gt;-2&lt;/sup&gt;)</td>
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<tr>
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<td>-14.0 ± 4.8</td>
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<td>19.2 ± 1.7</td>
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<tr>
<td>2017</td>
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<td></td>
<td>8.2 ± 2.9</td>
<td>11.6 ± 4.04</td>
<td>0.26 ± 0.08</td>
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<td></td>
<td>11.6 ± 4.04</td>
<td>0.37 ± 0.13</td>
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Table III. Comparison of SWARF 24 h at TOA with previous studies. These results were calculated for the biomass burning aerosols.

<table>
<thead>
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<th>Reference</th>
<th>Region</th>
<th>Period</th>
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</tr>
<tr>
<td>Sena et al. (2013)</td>
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<tr>
<td>Patadia et al. (2008)</td>
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<tr>
<td>Procópio et al. (2004)</td>
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<td>This work</td>
<td>Forest</td>
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</table>

*AF: Alta Floresta AERONET site.
**AH: Abracos Hill (Ji Paraná) AERONET site.**
Figure 1. a) Location of the study area. The yellow marks are T0a and T0e sites. The red mark is Manaus in the State of Amazonas, Central Amazon, Brazil, b) micrometeorological characterization for the T0a, showing the monthly averages for precipitation (PRP), air temperature (Tair), relative humidity (RU), and shortwave radiation.
Figure 2. a) Time series for aerosol optical depth (AOD 500 nm) retrieved by AERONET sunphotometer in Central Amazonia "T0a" ATTO, b) "T0e" EMBRAPA and c) linear regression for AOD 500 nm "T0e" and AOD 500 nm "T0a" instantaneous measurements.
Figure 3. a) Monthly distribution of fires over the legal Amazon and the states of Amazonas and Pará, Reference satellite Aqua MODIS afternoon (INPE), b) total AOD 500 nm, coarse and fine fractions (T0e), c) Angstrom exponent 440-870 nm (T0e), d) Precipitable water (T0e), e) Absorption coefficient 637 nm (T0a), f) Scattering coefficient 637 nm (T0a) and g) SSA 637 nm (T0a).
Figure 4. a) Boxplot for light scattering coefficient at 637 nm in T0a from 2014 to 2017, b) light absorption coefficient at 637 nm in T0a from 2014 to 2017, and c) for the single scattering albedo (SSA) at 637 nm in T0a “black” and T0e “red” from 2014 to 2017.
Figure 5. 24-hour compilation of fire spots, Aqua MODIS afternoon (INPE) reference satellite with the Hysplit trajectory model (backward trajectories) 04 Oct 2015.
Figure 6. Spectral variation of a) AOD, b) SSA and c) AP in the dry (red) and wet (blue) seasons in Central Amazonia (T0e) from 2011 to 2017. The lines represent the medians and intervals are 25 and 75 percentiles.
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