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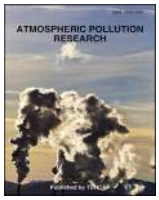

Optical properties and spectral dependence of aerosol light absorption over the Brazilian Pantanal

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ABSTRACT

Characterizing optical properties of aerosols, particularly the absorption processes, is fundamental for understanding the role of these particles in ecosystems as well as the climate, in general. Currently, changes in precipitation regimes in the southern Amazon basin have resulted in a considerable increase in biomass burning, whereby large amounts of aerosols and gases are emitted into the atmosphere. These increased emissions can impact the ecosystem, modifying mass and energy flows at the surface. This study, motivated by the need for more aerosol observations was the result of a long-term campaign carried out in the Brazilian Pantanal, which provided continuous in-situ measurements of aerosol optical properties between January 2017 and December 2019. From these data, optical properties of aerosols were quantified and analyzed seasonally. Corrected estimates for absorption coefficient (σ_{abs}) and Angstrom absorption exponent (α_{abs}) were utilized to characterize the spectral dependence of absorption by aerosols. With additional measurements of the scattering coefficient (σ_{scat}), it was possible to evaluate the single scattering albedo (ω_0). The mean values for σ_{abs}, σ_{scat}, ω_0 at 525 nm and α_{abs} 370–880 nm during the wet season were 0.66 ± 0.58 Mm^{-1}, 6.16 ± 5.75 Mm^{-1}, 0.90 ± 0.06 and 1.43 ± 0.49, respectively. These values, within measurement uncertainties, were similar to results found in the central Amazon. During the dry season, absorption and scattering increased approximately 88% and 86%, respectively, providing strong evidence for the contribution of local and regional emissions from biomass burning. The values of ω_0 did not reveal significant seasonal differences, however, a sharp reduction was observed, medians below 0.8, at the beginning of the burning period, which was associated with more recent local burnings and, therefore, greater absorption. The mean values evaluated for different spectral intervals reveal a strong contribution of brown carbon (BrC) during the dry season, with medians of 1.78 and 0.87 for the intervals of 370–590 nm and 590–880 nm, respectively. This work also quantified BC concentrations for the dry and wet seasons, obtaining mean values of 0.75 ± 0.83 and 0.12 ± 0.09 μgm^{-3}, respectively. In general, a striking similarity was encountered for aerosol optical properties between the Pantanal and the Central Amazon during the wet season. The results presented here characterize the background values for aerosol optical properties in the Pantanal, and the additional effects resulting from biomass burning emissions, therefore providing essential information to assess the effects these particles have on the ecosystem.

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1. Introduction

The radiative effects of atmospheric aerosols represent one of the greatest uncertainties in the global radiation budget (IPCC, 2013). While scattering of radiation due to aerosols causes a net cooling effect, aerosol absorption, in turn, contributes to the warming of the atmosphere (Rizzo et al., 2011). Aerosols that have strong absorption capacity are mainly composed of substances consisting of carbon and mineral dust. Soot, known as black carbon (BC), is almost purely made of carbon (Andreae and Gelencsér, 2006) and can strongly contribute to the absorption of solar radiation. BC emission results from various combustion processes, including biomass burning, and affects the terrestrial climate system directly through radiation absorption, and indirectly through changes in cloud properties (Bond et al., 2013; Malavelle et al., 2019; Morgan et al., 2019).

Although direct BC emissions are not soluble in organic solvents, such as water, soluble substances can accumulate on the BC particle surface, thereby increasing its potential to act as a cloud condensation nucleus (CNN) (Johnson et al., 2004; Sadiq et al., 2015). This particular aging process changes the lifetime and the BC removal rate, which makes it difficult to ascertain the precise role of this constituent in the climate system (Bond et al., 2013). In general, aerosols cool the climate system and, consequently, have an opposite effect to greenhouse gases; however, BC has a positive direct radiative forcing ($+0.6\ Wm^{-2}$) second only to CO_2 with respect to global warming (IPCC, 2013). Aerosol emissions from biomass burning, and particularly BC emissions, affect surface energy fluxes and atmospheric stability (Zhang and Wang, 2011; Thornhill et al., 2018, Liu et al., 2020). These emissions impact the overall radiative balance, directly influencing the aerosol-cloud and aerosol-radiation interactions. Furthermore, they also impact forest productivity, altering the concentrations of gases such as ozone and carbon dioxide (Aragão et al., 2018; Malavelle et al., 2019; Morgan et al., 2019; Neto et al., 2019; Liu et al., 2020; Pope et al., 2020), in addition to causing serious risks to human health (Andrade Filho et al., 2013; Galvão et al., 2017).

In addition to soot, emitted in biomass burning, there are also other types of carbonaceous radiation-absorbing aerosols, such as brown carbon (BrC), humic-type substances and biogenic aerosols (Andreae and Gelencsér, 2006; Sun et al., 2007; Laskin et al., 2015). Even though they are aerosols with absorption properties, BC and BrC exhibit different absorption spectral dependencies. BC is more absorbent at wavelengths in the near-infrared and BrC absorbs more in the near-ultraviolet range (Laskin et al., 2015). Based on the different absorption ranges, it is possible to differentiate these forms of aerosols through the spectral dependence of absorption, that is, through the Angstrom exponent of absorption (α_{abs}). For BC, its value is approximately 1.0 ± 0.1 for particles ranging from 10 to 100 nm in diameter (Sun et al., 2007; Gyawali et al., 2009). Some measurements taken in urban areas are consistent with this value range (Schnaiter et al., 2003; Bond and Bergstrom, 2006; Marley et al., 2009). However, field measurements for α_{abs}, during the burning season in the Amazon vary between 0.94 and 1.70 depending on the emission age and the estimation method (Rizzo et al., 2013; Saturno et al., 2018).

In Brazil, several studies have already characterized the absorptive properties of aerosols for the Amazon Rainforest (Schmid et al., 2006; Rizzo et al., 2011, 2013; de Sá et al., 2018; Saturno et al., 2018). However, to date, studies characterizing the optical properties of aerosols with in-situ, field measurements over the Brazilian Pantanal are non-existent. The Pantanal is one of the largest wetlands in the world and covers a total of 179.300 km^2 divided between Brazil, Bolivia and Paraguay, of which 140.000 km^2, approximately 78%, are in Brazil (Alho and Silva, 2012) divided in the states of Mato Grosso (MT, 35%), and Mato Grosso do Sul (MS, 65%) (Tomas et al., 2020). Due to its diversity and abundance of natural resources, local and regional economies highly depend on this biome (Padovani, 2017). Over the southern Amazon Basin and the Pantanal, modulation of the precipitation regime has led to more extreme droughts increasing the risk of fire (Marengo et al., 2018, 2021) which also drastically modifies the natural landscape (Ivory et al., 2019).

The Pantanal provides an excellent natural laboratory for monitoring the optical properties of aerosols, given that the wet season can be representative of a practically clean atmosphere that would be expected in undisturbed natural conditions. During the dry season, as in other areas of South America, the Pantanal is strongly influenced by emissions from biomass burning (Sena et al., 2013; Artaxo et al., 2013; Palácios et al., 2020; Marengo et al., 2021). Local studies employing remote sensing measurements have characterized the seasonal characterization of aerosol optical properties in the Pantanal (Palácios et al., 2016a; 2016b). Local in-situ measurements in the Pantanal of the absorption spectrum and scattering of radiation by aerosols can quantify the background values in the wet season and help to understand the effect of biomass burning emissions on this biome.

In this study, unique in-situ data was used to characterize the seasonal variation of optical properties of aerosols over the Brazilian Pantanal region. Using a corrective methodology with local environmental parameters for the absorption spectrum, optical values were obtained for both the wet season (background) and dry season (impacted by local and regional biomass burning). Average values for optical properties found for the Pantanal were used compared with representative values from Amazon Forest sites.

2. Methodology

2.1. Study area

In this study, data collected from a long-term campaign experimental site (January 2017 to December 2019) in the Pantanal (Fig. 1). The sampling site is located in the Baía das Pedras Park, within the Private Natural Heritage Reserve – RPPN SESC (16°39′ S; 56°47′ W) in the Poconé municipality and bordering the municipality of Barão de Melgaço (about 160 km from the capital Cuiabá) in the State of Mato Grosso, Brazil. This Pantanal region is considered one of the largest sedimentation plains on Earth (Fantin-Cruz et al., 2011; Paz et al., 2016). The biome is composed of a mosaic of landforms, with diverse vegetation species, with more than 1000 species of plants (Junk et al., 2011, 2014). Vegetation at the sampling site is monodominant, specifically *Combretum lanceolatum* Pohl, whose canopy height is on average close to 4 m (Nunes da Cunha et al., 2007).

In this work, the classification for dry and wet seasons was not established as a function of meteorological parameters; instead, the period in which the optical properties of aerosols change due to emissions from fires was assumed as the dry season. A similar classification was made by Rizzo et al. (2013) for the Amazon. The months from July to October were here called the dry season and the rest of the year as the wet season. Although the month of October already has a considerable rate of precipitation (see Fig. 3), records of local and regional burns still persist. The same can be interpreted for the month of July, in this case, although rainfall records are low, in this period the fires are still relatively low.

2.2. Instrumentation and measurements

Equivalent BC concentration measurements (hereafter referred to as simply BC) were obtained with a 7-channel model AE33 aethalometer (lacking the dual spot capacity) (λ = 370, 470, 520, 590, 660, 880 and 950 nm). Scattering measurements were performed with an Ecotech M9003 nephelometer with $\lambda = 525$ nm.

The aethalometer and nephelometer were installed inside the Pantanal Advanced Research Base with controlled temperature and relative humidity (air conditioning set at 25 °C and an air dehumidifier). This equipment was connected in parallel with a PM 2.5 cut-off section inlet at a 3 $Lmin^{-1}$ flow rate, with 2 $Lmin^{-1}$ being sampled by the aethalometer

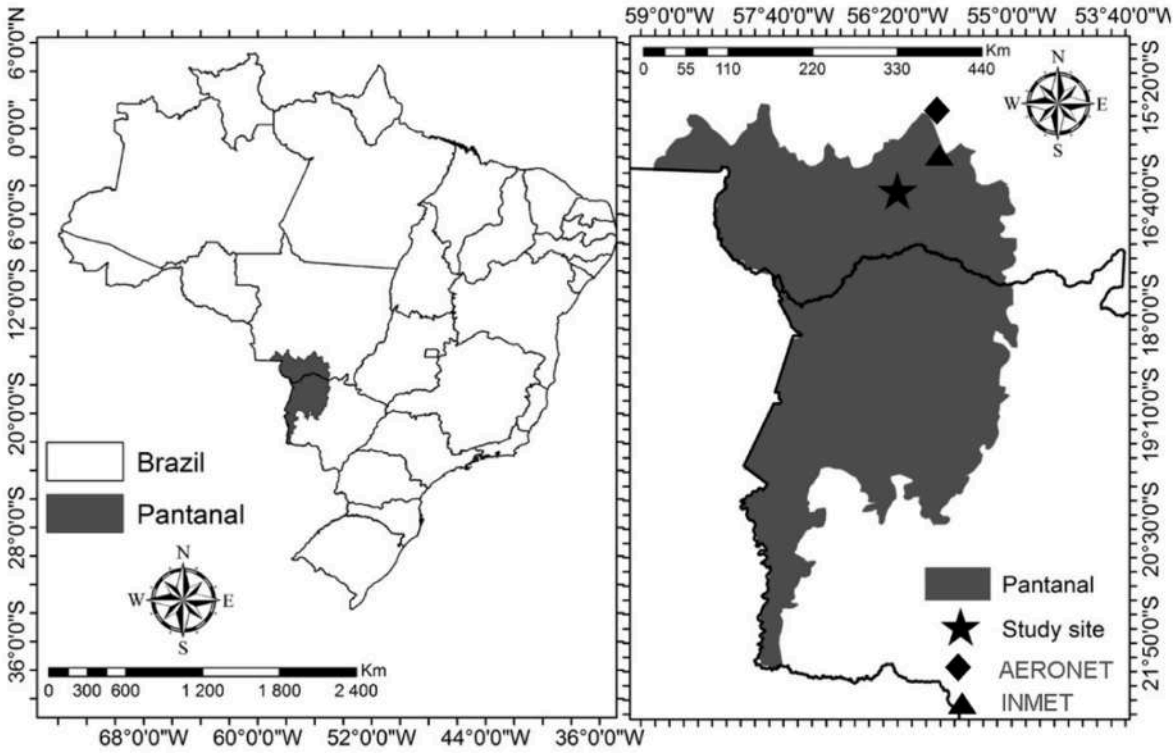

Fig. 1. Study site area in the Reserva Particular do Patrimônio Natural – RPPN SESC (16°39′ S; 56°47′ W). The Figure also shows the location of sites where complementary measurements were acquired: the AERONET CUIABA-MIRANDA site and the INMET micrometeorological site.

and 1 $Lmin^{-1}$ by the nephelometer. The inlet was installed at a height of 4.5 m above ground-level. Silica gel was used to control the humidity of the air sampled by the inlet. The Silica gel was removed and replaced weekly in order to keep sample air humidity below 40%.

BC concentration measurements (internally computed by the instrument using the 880 nm absorption coefficient) were performed every 10 min and scattering coefficient measurements every 5 min. The data analyzed here were obtained by calculating the average of these measurements every 30 min, corrected for standard temperature and pressure (*STP,* 273.15 *K* and 1013.25 hPa). To calculate the simple scattering albedo (ω_0) the absorption coefficient (corrected) at 525 nm was interpolated. To ensure the quality of the measured absorption spectral, a filtering criterion was used. All data whose BC concentration measurement were below the instrument's detection limit were excluded from the series (Rizzo et al., 2011).

Due to instrumentation limitations, measurements of absorption coefficients needed to be corrected (Arnott et al., 2005; Schmid et al., 2006; Rizzo et al., 2011). The correction method using local parameters and the possible errors for the absorption coefficients are shown in section 2.4 and in the supplementary material S1 and S2. Measurements for the scattering coefficient (measured by the nephelometer) are also subject to angular truncation errors. Based on Müller et al. (2011) and Rizzo et al. (2013) we can say that these errors can cause an underestimation of around 10%–20% in the scattering coefficients. As our instrument makes measurements at a single wavelength (525 nm), without backscattering measurements, it was not possible to use a correction model, so the measurements for the scattering coefficients are subject to this range of uncertainty.

2.3. Calculation of attenuation coefficients

Aerosol radiation attenuation coefficients were obtained from BC concentration data. The manufacturer's standard output gives the BC mass concentration (ngm^{-3}) that is calculated internally by the instrument using the attenuation coefficient, (Equation (1)):

$$BC = \frac{\sigma_{atn}}{\delta_{atn}}, \tag{1}$$

where:

$$\delta_{atn} = 14625/\lambda \ \left[m^2 g^{-1}\right]. \tag{2}$$

In Equation (2), δ_{atn} the optical absorption cross-section is based on a calibration at a wavelength of 880 nm using the Malissa-Novakov method, a solvent-based thermal desorption method for elemental carbon analysis (Gundel et al., 1984). Equations (1) and (2) were used to convert the BC concentration into the values of σ_{atn}, (used in calculating the absorption coefficients), following the studies of Schmid et al. (2006), Rizzo et al. (2011) and Zhuang et al. (2015).

It is worth noting that both the absorption and scattering coefficient decrease with the wavelength following a power law, where:

$$\sigma_{(\lambda)} = A \ \lambda^{-\alpha}, \tag{3}$$

being α the Angstrom exponent which can be calculated for an interval of according to Equation (4):

$$\alpha = -\left[\frac{\ln\left(\frac{\sigma_{(\lambda_1)}}{\sigma_{(\lambda_2)}}\right)}{\ln\left(\frac{\lambda_1}{\lambda_2}\right)}\right]. \tag{4}$$

Thus, Equation (4) can be used to calculate both the absorption Angstrom exponent (α_{abs}) whose absorption coefficients are $\sigma_{abs\ (\lambda_1)}$ and $\sigma_{abs\ (\lambda_2)}$ as well as to calculate the scattering Angstrom exponent (α_{scat}) whose scattering coefficients are $\sigma_{scat\ (\lambda_1)}$ and $\sigma_{scat\ (\lambda_2)}$.

2.4. Obtaining absorption coefficient

The attenuation coefficient (σ_{atn}) can differ significantly from the aerosol absorption coefficient (σ_{abs}) due to optical interactions of the

filter substrate with the deposited aerosol (Petzold et al., 1997; Weingartner et al., 2003; Arnott et al., 2005; Schmid et al., 2006; Moosmüller et al., 2009; Müller et al., 2011). For these cases, the absorption coefficient is obtained by correcting the values of σ_{atn} (Equation (5)):

$$\sigma_{abs} = \sigma_{atn}\frac{1}{C.R_{atn}}, \quad (5)$$

where $C(\geq 1)$ is the multiple scattering effect and depends mainly on the nature of the filter and the device used; and R_{atn} explains the filter loading effects, varying with the amount of aerosol particles entering the filter and their optical properties.

The aerosol load correction of the filter depends on the attenuation measured directly by the aethalometer (σ_{atn}) and can be calculated using the following empirical relationship (Equation (6)):

$$R_{atn} = \left(\frac{1}{f} - 1\right)\frac{\ln\ \sigma_{atn} - \ln\ 10}{\ln\ 50 - \ln\ 10} + 1, \quad (6)$$

where the parameter f is known as the shading factor and depends on the optical properties of the aerosol type. For values $\sigma_{atn} \leq 10$ of the aerosol load is so low that aethalometer performance is not affected $(R_{atn} = 1)$ (Weingartner et al., 2003; Schmid et al., 2006). On the other hand, for, $\sigma_{atn} > 10$ begins to decrease with $R_{atn}\sigma_{atn}$.

According to Equation (6), for $f = 1$, $R_{atn} = 1$ is independent of σ_{atn}; if $f > 1$, R_{atn} it becomes smaller than unity. As such, f can be described as a shading parameter or factor. Schmid et al. (2006) obtained $f = 1.2$ (when the simple scattering albedo, $\omega_0, \approx 0.9$) using empirical methods and suggested that this value is wavelength-independent. As in other works (Weingartner et al., 2003; Schmid et al., 2006; Zhuang et al., 2015), $f = 1.2$ was assumed in this study.

The multiple scattering effect (C) is by far the most important effect when inferring values σ_{abs} as a function of σ_{atn} and can be calculated for each wavelength using Equation (7) (Schmid et al., 2006):

$$C(\lambda) = C^{*}(\lambda) + m_s(\lambda)\frac{\omega_0(\lambda)}{1 - \omega_0(\lambda)}, \quad (7)$$

where m_s represents the aerosol scattering coefficient fraction that is misinterpreted as absorption, $C^{*}(\lambda)$ is the reference multiple scattering factor that includes the aerosol scattering effects, ω_0 is the aerosol single scattering albedo, and λ is the wavelength in nm. The spectral dependence of C was estimated considering a power law (Equation (4)) whose parameters have been adjusted through a quadratic fit for a range of values of σ_{abs} between 1.0 and 2.6. The values of σ_{abs} were then retrieved by an iterative procedure, similar to that performed by Schmid et al. (2006) and Rizzo et al. (2011), until α_{abs} converged to a precision of 5%. The full description of corrections/compensations and sensitivity tests can be found in the Supplementary Material (section S.1). Here, typical uncertainties were from ±4% for σ_{abs} and ±17% for α_{abs} considering the entire study period. It is reasonable to consider these uncertainties for all points, since the days chosen for the sensitivity test (Supplementary Material, S.2) represent the critical period of the year, when the highest records of local and regional biomass burning occur.

2.5. Complementary measurements

To complement the discussions related to variations in optical properties, this work used the records of fire outbreaks from the National Institute for Space Research, INPE (INPE, 2020) (https://queimadas.dgi.inpe.br/queimadas/portal). The measurements from the *Aqua* reference satellite (afternoon period) were used for the entire Legal Amazon, in addition to the records for the State of Mato Grosso and for the Pantanal biome. Monthly accumulated figures for fire outbreaks from January 2010 to December 2019 were obtained, in addition to local measurements for the municipalities of Poconé and Barão de Melgaço for the period 2017–2019. Precipitation measurements (PPT) were obtained by the National Institute of Meteorology, INMET (INMET, 2020) (https://mapas.inmet.gov.br/). Data from the conventional meteorological station Padre Ricardo Remetter, identification 83364, were used. Station 83364 is approximately 60 km (straight line) from our site in the Pantanal. The monthly accumulated PPT for the years 2017–2019 was used. The CUIABA-MIRANDA site of the AERONET network (Holben et al., 1998) was also used to obtain the optical depth for the fine fraction of the aerosol (Fine Mode AOD 500 nm). Version 3 data was obtained: SDA Retrieval Level 2.0. The data acquired for the period 2017–2019 were used to calculate monthly averages, analyzed in order to verify the variations of measured properties in the Pantanal.

3. Results and discussion

3.1. Correction factors

The correction factors for the effects of multiple scattering, $C(\lambda)$, and filter loading, R_{atn}, for Pantanal atmospheric conditions are shown in Table 1. The average value of C at 660 nm was 6.02 for the entire sampling period, a value similar to that found by Rizzo et al. (2011) for the Amazon ($C = 5.72 \pm 0.14$ at 660 nm). In the work by Schmid et al. (2006), values of $C = 4.97$ to 6.73 for $\lambda = 450$–950 nm were obtained for the SMOCC experiment in Amazonia.

In comparison with other works (e.g., Collaud Coen et al., 2010; Ran et al., 2016), our results approximate relatively well with the results of Schmid et al. (2006) and Rizzo et al. (2011) developed during campaigns in the Amazon, which suggests that aerosols in the Pantanal have similar characteristics to Amazon aerosols. The standard deviations in Table 1 are small, indicating that $C(\lambda)$ varied little during the study period, even with different atmospheric conditions experienced during the dry and wet seasons. The values of R_{atn} practically did not vary with wavelength. Furthermore, (R_{atn} ~1.0) values close to one (1.0) suggest that for less polluted atmospheres, such as that of the Pantanal, the loading effect on the filter is minimal.

Regarding corrected σ_{abs} and α_{abs} values, differences were more notable during the dry period (July to October). The σ_{abs} 590 nm values were, on average, approximately 82% lower than the corresponding attenuation coefficients. The Angstrom Absorption, α_{abs} 370–880 nm was approximately 36% larger than the corresponding Angstrom attenuation exponents.

3.2. Aerosol optical properties variability

Daily averages for the entire time series of the scattering (σ_{scat}) and absorption (σ_{abs}) coefficients are illustrated in Fig. 2. Although there are annual variations, seasonal variation is evident, where maximum values for each year are encountered during the dry season. This seasonal pattern was not expected as a consequence of the higher concentration of fine mode particles in the dry season, as verified by Palácios et al. (2020) for the AERONET site of Cuiaba-MIRANDA. Similar results were obtained for sites in the Amazon (Artaxo et al., 2013; Rizzo et al., 2013; Saturno et al., 2018). Rizzo et al. (2013) also state that the biomass burning emission has a greater efficiency in radiative scattering when compared to the biogenic emission, predominantly in the aerosol coarse

Table 1
Correction factors spectral variation due to effects of multiple scattering $C(\lambda)$ and filter loading R_{atn} calibrated for the atmospheric conditions of the Pantanal.

λ (nm)	$C(\lambda)$	R_{atn}
370	4.090 ±0.003	1.016 ±0.019
470	4.810 ±0.001	1.018 ±0.048
520	5.150 ±0.001	1.020 ±0.059
590	5.600 ±0.001	1.020 ±0.064
660	6.020 ±0.004	1.020 ±0.068
880	7.240 ±0.014	1.030 ±0.072
950	7.600 ±0.017	1.040 ±0.083

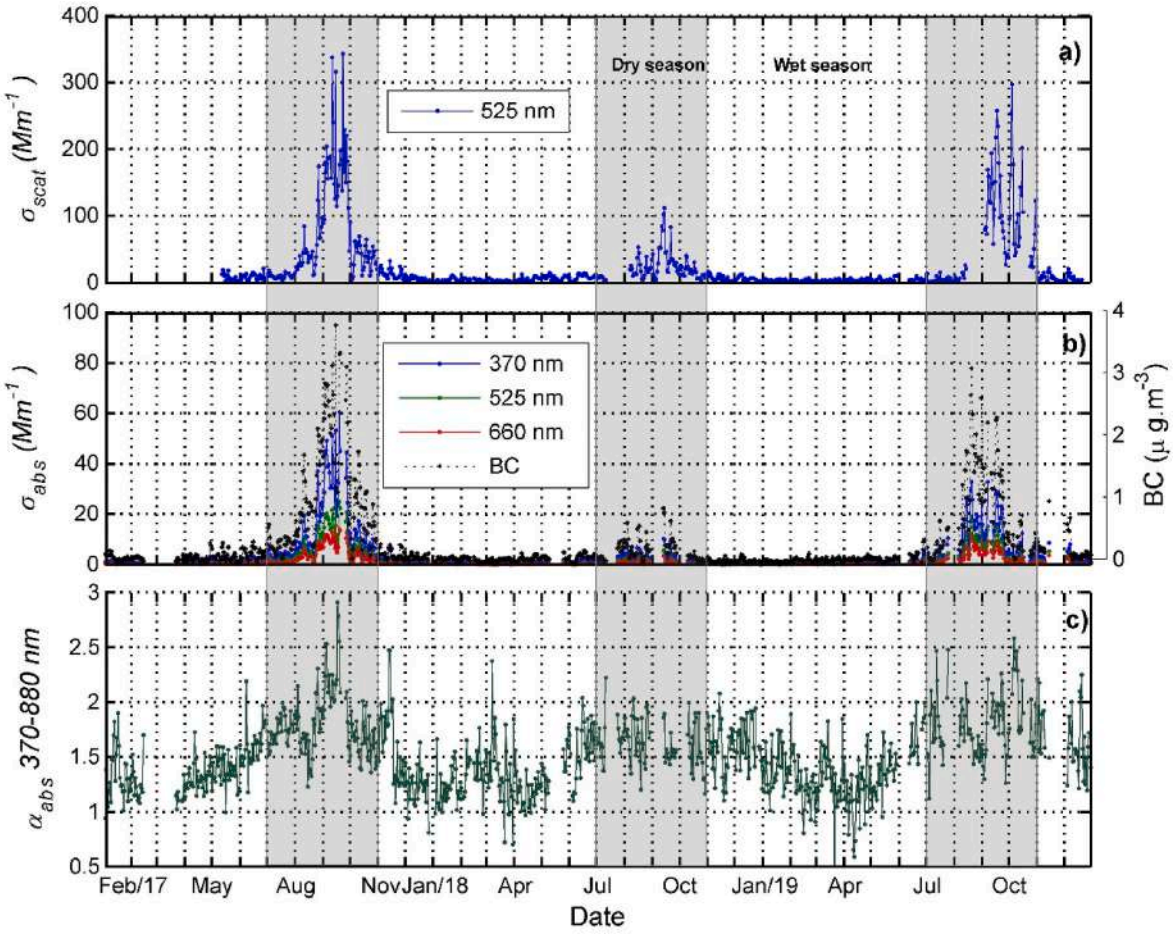

Fig. 2. Daily mean for time series, a) scattering coefficients (σ_{scat}) at 525 nm, b) absorption coefficients (σ_{abs}) at 370, 525 and 660 nm and black carbon concentration (BC), in μgm^{-3}, c) Angstrom exponent of absorption (α_{abs}) between 370 and 880 nm. Calculations were based on 30 min values. Shaded areas represent the dry season.

mode.

With Fig. 2, it is also possible to observe the difference between σ_{scat} and σ_{abs} for the 2018 dry season when compared to the years 2017 and 2019. The median of σ_{scat} at 525 nm for the 2017 and 2019 dry seasons were higher by approximately 50% and 53%, respectively, the 2018 dry season. This may result from occurrence of local and regional fire outbreaks over the Pantanal. The work by Rizzo et al. (2013) found annual differences of up to 110% for scattering in the Amazon, a difference consistent with forest fire records. Descriptive statistics for the seasonal and annual variations of the scattering, absorption and simple scattering albedo coefficients are shown in Table S1 (Supplementary Material).

Absorption coefficients (σ_{abs}) for wavelengths 370, 525 and 660 nm are shown in Fig. 2b. Measurements of σ_{abs} for different wavelengths show an increase in the magnitude of absorption towards the ultraviolet. Studies in the Amazon such as those by Rizzo et al. (2011) and Rizzo et al. (2013) have already reinforced that the shorter the wavelength, the greater the interaction with fine fraction aerosols and the greater the scattering and absorption coefficients. Studies carried out in urban regions have also verified these relationships. Mogo et al. (2012) found approximate differences of 15% between σ_{abs} at 470 nm and 522 nm and 32% between 470 nm and 660 nm in an urban region in central Portugal. For comparison purposes, average differences found between σ_{abs} at 370 nm and σ_{abs} at 525 nm and σ_{abs} at 660 nm were approximately 50% and 65%, respectively. The proportions between absorptions at different wavelengths provide information about the physical characteristics of the aerosol population (Russell et al., 2010, Cazorla et al., 2013; Mogo et al., 2017; Palácios et al., 2020). Detailed discussion about the absorption spectrum is presented in section 3.4.

To assess the possible contributions of local and regional fire outbreaks, this work analyzed the historical records of fire outbreaks (INPE, 2020) for the entire Legal Amazon as well as those occurring in the State of Mato Grosso and specifically in the Pantanal biome. The time series (2010–2019) containing this information is shown in the Supplementary Material (Fig. S3). Local estimates were also obtained for fire outbreaks in the Poconé and Barão de Melgaço municipalities in the State of Mato Grosso, located near the experimental sites. These numbers indicate that the principle annual variations in the values of σ_{scat} and σ_{abs} in Fig. 2, may result from local and regional biomass burning. The verification of these sudden variations in the annual properties was also confirmed with the AERONET measurements at the Cuiaba-MIRANDA site as shown in Fig. 3.

The differences in the optical depth of the fine-mode aerosols (fine AOD) for September between 2017 and 2019 in relation to 2018 directly follow the variations of fire outbreaks in the Pantanal. It was found that the fine AOD (September) for the years 2017 and 2019 were approximately 70% and 75%, respectively, higher than the values for 2018. The work by Marengo et al. (2021), which discusses extreme Pantanal droughts that occurred in 2019 and 2020, emphasizes that since 2019 the Pantanal has suffered a prolonged drought that has had extreme consequences for the region. These authors also found that the lack of rain in the summers of 2019 and 2020 over the region was caused by the reduction in the transport of hot and humid air from the Amazon to the Pantanal, which triggered subsequent biomass burning events that involved hundreds of thousands of acres.

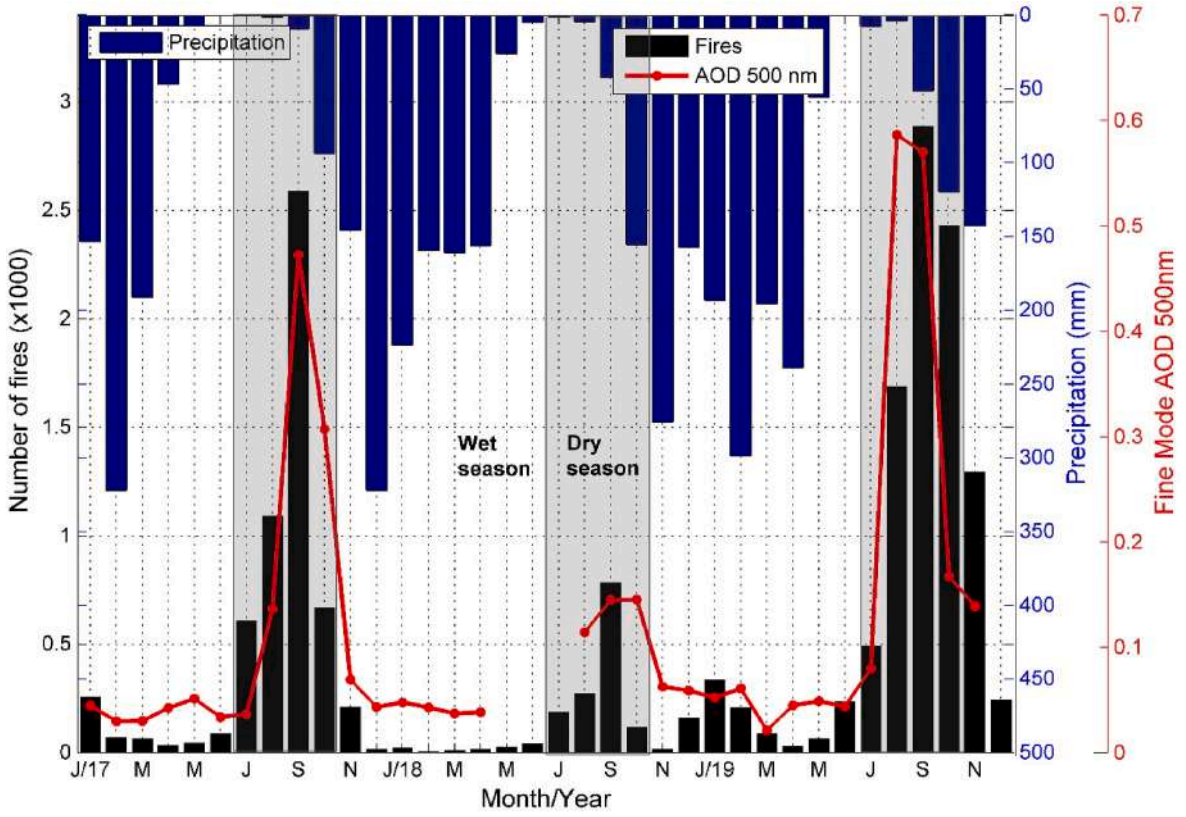

Fig. 3. Relationship between the monthly rainfall accumulated, the monthly number of the Pantanal fires and the aerosol optical depth in fine mode (fine mode AOD 500 nm). The shaded areas represent the seasons considered to be dry in this study.

Fig. 2 b) contains the time series for black carbon concentration (BC), in μgm^{-3}, obtained for the Pantanal. Evidently, the variations in concentrations followed the variations in the absorption coefficients (Shown in Table S1, Supplementary Material). For comparison purposes, the study of Santos et al. (2016), carried out over the Pantanal (2012–2013) with filter analysis experiment campaign, found a maximum BC concentration in fine mode of 1.68 μgm^{-3} in September 2012, a result approximately 2.5 times lower than that obtained for September 2019 in this work. Since BC emission is mostly soot from combustion, this difference may be linked to the greater number of fires that occurred locally and regionally in 2019 (Marengo et al., 2021).

In contrast to this result, Artaxo et al. (2013) showed that on a site highly impacted by biomass burning in the Amazon, the Porto Velho site, the maximum BC (fine mode) values can reach 22 μgm^{-3}, that is, a difference of approximately 80%, when compared to Pantanal maximum. BC concentrations in the Pantanal compared with Porto Velho (2009–2012), the BC averages for the dry and wet seasons of Porto Velho were higher by approximately 73% and 70% for the respective dry and wet seasons in the Pantanal. It is evident from these comparisons that sites directly impacted by biomass burning in the Amazon, such as the case of Porto Velho, are much more impacted by the concentration of BC than the Pantanal, which is mainly affected by local and regional emissions from biomass burning.

When comparing the BC concentration averages between the Pantanal and the ZF2 site (2008–2012), central Amazonia (Artaxo et al., 2013), differences were found between the dry and wet seasons of approximately 55% and 15%, respectively, values for the Pantanal being higher. Within the uncertainties, there is practically no difference between these concentrations for the wet season. This result lends credence to our statement that the Pantanal experiences, during the wet season, an atmosphere nearly as clean as the atmosphere of a preserved forest, in terms of BC concentration. As for the dry season, it can be observed that the influence of local and regional fires increases BC concentrations in the Pantanal, causing their concentrations to be higher than regional emissions that affect the central Amazon.

The monthly variations of aerosol optical properties in the Pantanal are illustrated in Fig. 4. The boxplots for the scattering coefficients (σ_{scat}), absorption (σ_{abs}) and for the single scattering albedo (ω_0) were calculated for 10 Julian Day intervals between January 2017 to December 2019. The seasonal variation for the scattering and absorption coefficients was more evident (also shown in Fig. 2 and supplemented in Table S1). For ω_0 the median values for the month of June vary from 0.88 to 0.92, however, for the month of July these values vary between 0.76 and 0.84; that is, they experience a maximum variation of up to 17%. This greater variability in values ω_0 accompanies the intense activity of local biomass burning.

When analyzing the fire outbreaks records for the municipalities of Poconé and Barão de Melgaço, it was found that, on average, the monthly fire occurrence rises from 24 outbreaks in June to 80 in July, that is, they experience an increase of 70% right at the beginning of the dry season (INPE, 2020). Thus, the variations in the values of ω_0 right at the beginning of the increase in the burning records show that the local burning, fresher burning, can emit more absorbing particles, changing the values of ω_0 (Reid et al., 1998). It is observed that the increase in fire reports in the following months, the values of ω_0 rise again, which may be a consequence of the mixture of new particles with aged particles emitted locally or even arising from atmospheric transport.

For Amazon sites the monthly behavior of ω_0 is different, however, the seasonal and total averages are similar. Table 2 shows a comparison between the average values of the main optical properties obtained in the Pantanal and the average values obtained for two sites in central Amazonia (Rizzo et al., 2013; Saturno et al., 2018). Although these comparisons indicate small differences in the measured spectral ranges, it is possible to observe similar mean seasonal behavior. Table 2 is

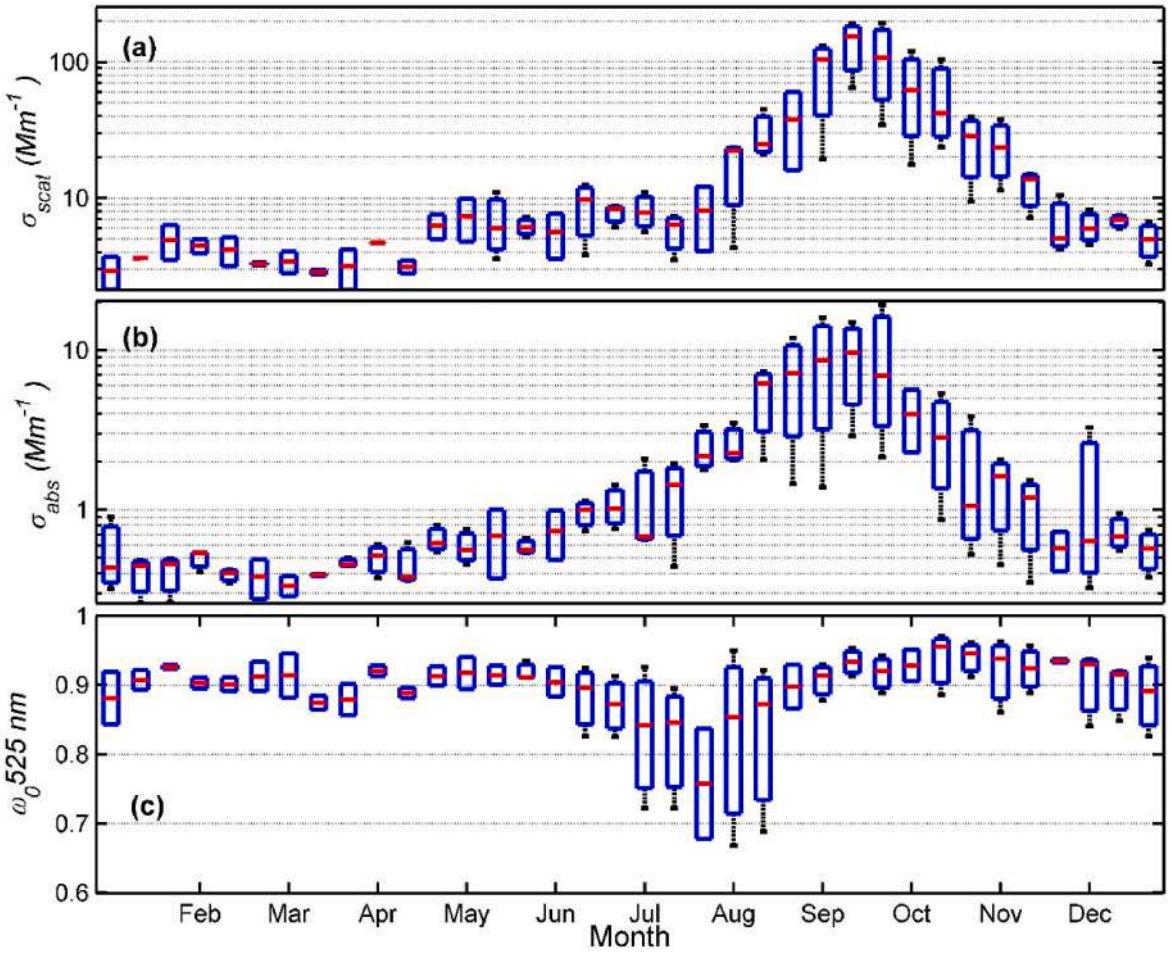

Fig. 4. Boxplot of a) scattering coefficients (σ_{scat} 525 nm), b) absorption coefficients (σ_{abs} 525 nm) and c) single scattering albedo (ω_0 525 nm) calculated for every 10 Julian days between May 2017 and December 2019. Boxplots calculated for of every 10 julian days of all three years of data combined.

Table 2
Comparison of mean seasonal values and for the campaign period for the main optical properties obtained for the Pantanal and for two sites in central Amazonia. Comparison with the results of Rizzo et al. (2013) on the ZF2 website and with the results of Saturno et al. (2018) on the ATTO website. The scattering coefficient (σ_{scat}) absorption (σ_{abs}) and single scattering albedo (ω_0) values were obtained at 525 nm for the Pantanal, while for ZF2 and ATTO σ_{scat} was obtained at 550 nm, σ_{abs} and ω_0 were obtained at 637 nm. The Angstrom exponent of absorption values (α_{abs}) were obtained over the 370–870 nm range for the Pantanal, 440–700 nm for the ZF2 and by way of a linear adjustment for the ATTO.

	Rizzo et al. (2013)			Saturno et al. (2018)			This work		
	ZF2 (2008–2011)			ATTO (2012–2017)			Pantanal (2017–2019)		
	Wet	Dry	All	Wet	Dry	All	Wet	Dry	All
σ_{scat} (Mm^{-1})	8.1 ± 7.2	36.0 ± 48.0	21.0 ± 36.0	7.5 ± 9.3	33.0 ± 25.0	22.0 ± 25.0	6.16 ± 5.75	54.51 ± 81.53	24.25 ± 55.27
σ_{abs} (Mm^{-1})	1.0 ± 1.4	3.9 ± 3.6	2.3 ± 3.0	0.68 ± 0.91	4.0 ± 2.2	2.1 ± 2.2	0.66 ± 0.58	4.73 ± 5.62	2.22 ± 4.03
α_{abs}	1.48 ± 1.12	1.70 ± 0.41	1.59 ± 0.86	0.91 ± 0.19	0.94 ± 0.16	0.93 ± 0.16	1.43 ± 0.49	1.75 ± 0.42	1.55 ± 0.48
ω_0	0.86 ± 0.09	0.87 ± 0.06	0.86 ± 0.08	0.93 ± 0.04	0.87 ± 0.03	0.89 ± 0.04	0.90 ± 0.06	0.90 ± 0.07	0.90 ± 0.06

further complemented with the mean values for BC concentration in the Pantanal.

Within the limits of the standard deviations, the average values for the scattering in the Pantanal are equal to those of the central Amazon; however, the average value for the dry season is slightly higher. In the dry season, σ_{scat} for the Pantanal it was approximately 35% and 40% higher than the values obtained for the ZF2 and ATTO sites, respectively. The justification for this result can be associated with geographic location and possible contributions from emissions from other locations. The contribution of burning emission from forest, cerrado and pasture vegetation may have influenced the scattering and absorption values in the Pantanal (Palácios et al., 2020).

For the average absorption values for the Pantanal, no statistical difference was found when compared to those of the Amazon, however, it is possible to observe small variations. During the wet season, σ_{abs} for the Pantanal was approximately 35% lower than the result for ZF2, which can be explained by the contributions of African aerosols arriving in the Amazon during the wet season (Rizzo et al., 2013). There are still no reports that these contributions can reach the central region of Brazil and, therefore, it needs to be stated with caution that the Pantanal is free from these influences in the wet season. For the dry season, there is an inversion, the value of σ_{abs} for the Pantanal was approximately 17% higher than the value found for ZF2, a fact that can also be explained by the fresh burning that occur locally. The values obtained for the

Angstrom exponent of absorption (α_{abs}) are also comparable in magnitude to those found in ZF2 (Rizzo et al., 2013).

3.3. Diurnal cycle of aerosol optical properties

The diurnal cycle at hourly intervals of scattering (σ_{scat} 525 nm), absorption (σ_{abs} 525 nm), simple scattering albedo (ω_0 525 nm) and absorption Angstrom exponent (α_{abs} 370–880) are shown in Fig. 5. For the wet season, a regular behavior for all analyzed variables, the medians of σ_{scat} and σ_{abs} showed a behavior similar to that described by Rizzo et al. (2013). From 08:00am (LT) the values of σ_{scat} and σ_{abs} start to increase varying by approximately 34% until 12:00pm (LT). For the Amazon, Rizzo et al. (2013) found an increase as large as 50% and attributed this variation to the increase in submicron particle size, pointing out that these diurnal variations for larger sizes have already been observed at other Amazon sites and can be justified by the photochemical formation of secondary organic aerosols in the residual layer. These aerosols are mixed downward towards ground level during turbulent exchanges during early morning boundary layer growth.

The work by Saturno et al. (2018) also found similar variation for absorption during the wet season and attributed these variations with the development of the boundary layer. With complementary measures of concentration in the accumulation mode, Saturno et al. (2018) also showed that these concentrations follow the same absorption pattern. These results argue for the development of a stable nocturnal layer resulting in particle deposition in the canopy, decreasing their concentration during nighttime hours. With the incidence of solar radiation, in the early hours of the day, the vertical turbulent mixture breaks down nocturnal stability causing concentrations to rise again.

Due to the lack of complementary measures, and observing the pattern found for the hourly variations of scattering and absorption during the wet season, this work suggests that the hourly dynamics of aerosols in the Pantanal is consistent with the findings of Rizzo et al. (2013) and Saturno et al. (2018) in the Amazon. However, future work needs to be carried out in the Pantanal to better understand these hourly variations, without discarding, for example, the possible diurnal emission of biogenic particles, as indicated by Rizzo et al. (2013). During the wet season, the consequences of variations of σ_{scat} and σ_{abs} on the values of ω_0 and α_{abs} can be observed. The maximum values of ω_0 occur just after 10:00am as a possible consequence of the addition of scattering particles that had been deposited within the canopy (Saturno et al., 2018) or were formed by secondary organic aerosols (Rizzo et al., 2013). The values of α_{abs} vary little during daytime hours, however, an inverse behavior of the previous variables, there was an approximate difference of 13% between day to night, with the lowest values found for daytime hours. Rizzo et al. (2011) found an approximate 20% variation in a pasture site in Amazonia and also justified this difference as due to boundary layer dynamics, but also noting that it may be a consequence of particle size change.

For the dry season, the hourly behavior of σ_{scat}, σ_{abs}, ω_0 and α_{abs} is shown in Fig. 5 (a-c-e-g). Differently from the results of Saturno et al. (2018), hourly variations are not very pronounced for σ_{scat}, σ_{abs} and ω_0.

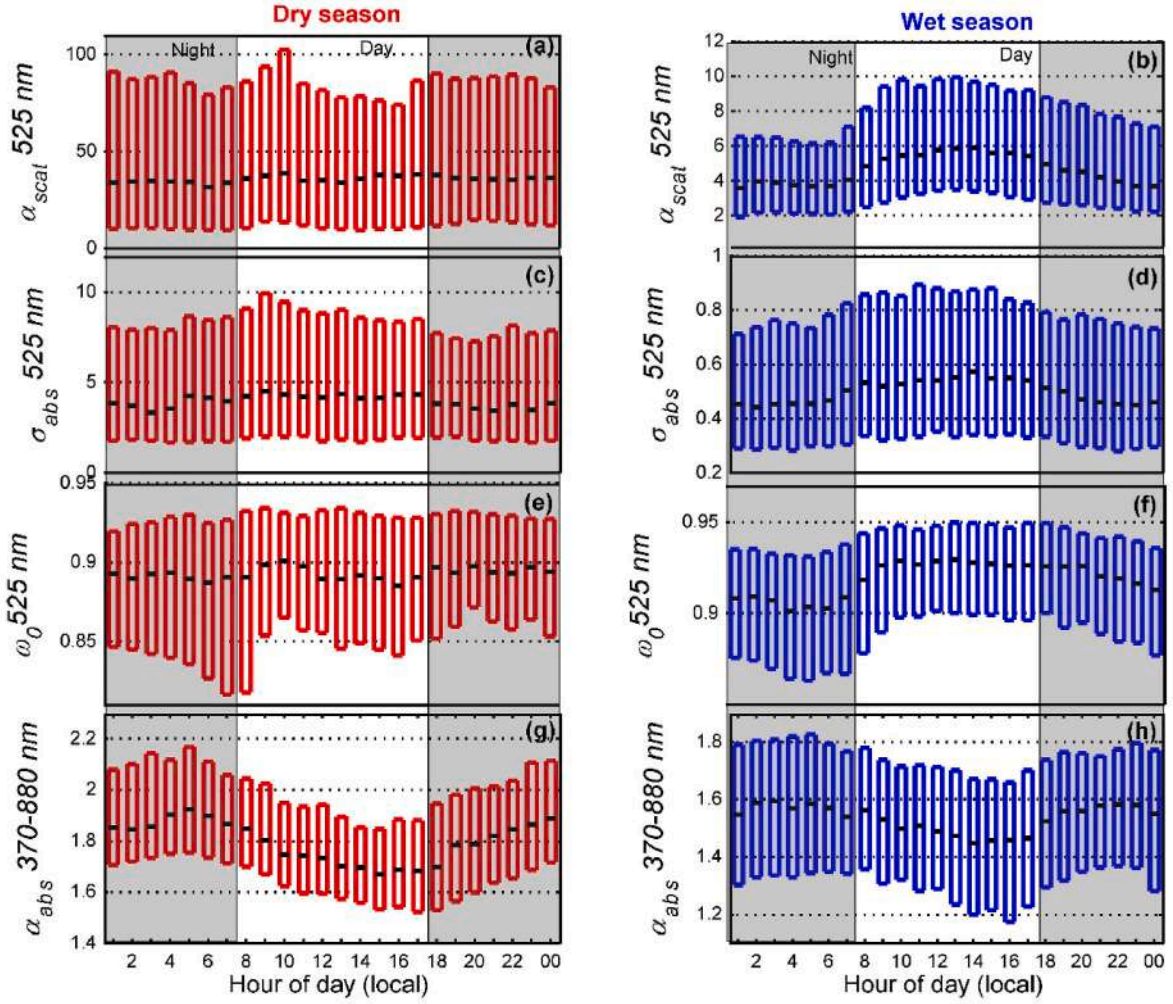

Fig. 5. Daily variation of (a, b) median scattering coefficient (σ_{scat} 525 nm), (c, d) median absorption coefficient (σ_{abs} 525 nm), (e, f) median simple scattering albedo (ω_0 525 nm), (g, h) median Angstrom exponent of absorption (α_{abs} 370–880 nm).

It is believed that the addition of emissions from local fires and also from regional transport can overload the natural dynamics of the aerosol (Rizzo et al., 2013), so that no pattern of time variation is defined. The variations of α_{abs} are well defined, with minimum values, during the day. Also, according to Rizzo et al. (2013) these variations are due to the diurnal contributions of regional emissions that are older. Burning emissions, when aged, undergo processes of coagulation and condensation of organic and inorganic species and change the size of the aerosol. For the Amazon region, which is intensively affected by emissions from fires, Chand et al. (2006) (SMOCC experiment) found that diurnal aerosols are more aged, justifying these findings by the contribution of regional emissions.

During the period considered as the dry season, the characteristic movement of the atmosphere contributes for plumes from the north, northeast and northwest directions to reach the study site, while in the wet season the movement is characteristically from the north with some isolated contributions from the south and southeast (masses of cold air). The local work of Palácios et al. (2018) performed backtrajectory simulations with Hysplit on the study site, highlighting the contributions of regional fires on the Pantanal.

3.4. Spectral dependence of aerosol light absorption

The average absorption spectra calculated for the experimental period (2017–2019) were subdivided by year and dry and wet seasons, as shown in Fig. 6. With the exception of 2018, the results show considerable differences between the curve adjustments for the dry and wet seasons. Variations between the dry and wet seasons were already expected, as the Pantanal, as well as other Brazilian biomes, is strongly influenced by burning during the dry season. As verified by Palácios et al. (2020), absorption spectra variability is clearly dependent on the records of fire outbreaks that occur in this state.

Absorption is also a function of particle size, the smaller the diameter, the greater the interaction of particles with shortwave radiation, resulting in greater absorption and deviations. Cut-off diameter diameter was PM 2.5, the resulting in total averages of σ_{abs} (2017–2019) were on the order of 4.15 ± 8.73 Mm^{-1}at 370 nm and 0.67 ± 1.11 Mm^{-1}at 950 nm. These results are well below those found by Zhuang et al. (2015) for the urban area of Nanjing, China (82.07 Mm^{-1}at 370 nm to 17.56 Mm^{-1}at 950 nm for 2012 and 2013 averages) which suffers from high intensity biomass burning and pollution episodes. The Pantanal, in general, suffers little influence from urban and industrial pollution, given its locate near a small town and large distances sizeable industrial centers.

In Fig. 6, absorption coefficients decrease by a factor of approximately 10 at all wavelengths when moving from the dry to the wet season. This fact can be directly associated with the decrease in aerosol concentration in the wet season. Evidently, with the reduction in concentration, mainly in the fine particle fraction, there is a change in the size distribution (Santos and Nogueira, 2015). In the supplementary material, Fig. S4 shows the change in size distribution from the wet to the dry season at AERONET's Cuiabá-MIRANDA site. As in Amazonian sites, a possible explanation for this reduction would be the removal of soluble organic aerosols during precipitation processes (Rizzo et al., 2011). This would result in a higher proportion of insoluble carbon present in the atmosphere. This proportion was also found for an undisturbed forest site in central Amazonia by Rizzo et al. (2013).

To assess the variability of the spectral dependence with respect to the mean values represented in Fig. 6, α_{abs} was computed for three pairs of wavelengths (370–590, 370–880 and 590–880 nm). These mean values, medians and total numbers of α_{abs} 370–880 and 590–880 utilized in the calculation are shown in the Supplementary Material in Table S1. The separation of α_{abs} for different pairs of absorption coefficients contributed to the analysis of the absorption processes. The fact that BrC absorbs strongly at short wavelengths, close to 440 nm (Wang et al., 2016), justifies the differences found between α_{abs} 370–590 nm and α_{abs} 590–880 nm. We believe that the median of α_{abs} 370–590 nm (1.78) for the dry season is a result of the strong contributions of BrC, whereas for α_{abs} 590–880 nm (0.87), free of BrC, it would be a result of the BC contribution. The work by Kirchstetter et al. (2004) points out, in a specific analysis of the absorption spectrum, that α_{abs} for the biomass burning emission is approximately 2. For the Pantanal, during the dry season the average value found was 1.81 ± 0.30 in the range 370–590 nm, being that of the total points of 370–590 nm, approximately 80%

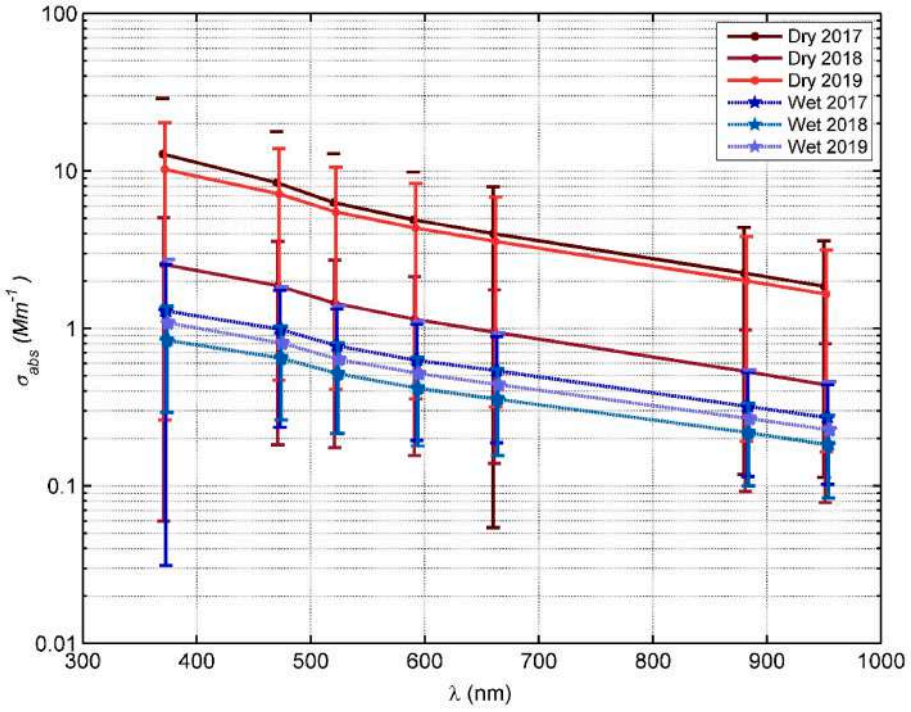

Fig. 6. Average aerosol absorption spectra for the years 2017, 2018 and 2019 for the dry and wet seasons. Error bars represent the standard deviation.

were between 1.5 and 2.5. These results agree with the analysis by Rizzo et al. (2011) for a pasture site in the Amazon.

Different absorptions in different bands of the spectrum suggest well-defined groups for aerosols in the dry and wet seasons. The strong absorption at 370 nm in the dry season (2017 and 2019) is indicative of aerosols formed by a complex mixture of incomplete burning of local biomass with contributions from regional particles transported to the study site. The magnitude of the absorption coefficients in the dry season together with the values of α_{abs} reinforce the contribution of organic material to the absorption in the spectral range close to the ultraviolet. The values of α_{abs} for 370–590 nm in Fig. 7 confirm the contribution of BrC in the absorption processes. In the rainy season, the chemical characterization work by Santos and Nogueira (2015) states that the main groups of aerosols in the Pantanal are formed by biogenic emissions, which explains the considerable reduction in absorption values.

Fig. 7 shows the histograms of relative frequency and normalized cumulative frequency for the different α_{abs}. The values of α_{abs} for the spectral ranges, 370–590 nm and 370–880 nm, (Fig. 7a, b, d, e) show a similar behavior with a shift to the right (higher values) in the dry season, as mentioned above, result of a possible increase in the concentration of fine particles emitted by biomass burning. The values of α_{abs} in the range 590–880 nm (Fig. 7c, f) show a similar behavior between the dry and wet seasons, with a difference of approximately 10%. Although there is no statistically significant difference between the medians for the dry and wet seasons, this small increase to values close to 1.0 in the dry season (median of 0.87) may be due to BC emissions.

For the wet season, the results of this work suggest that the contributions of biogenic emissions, free from biomass burning, are responsible for the reduction of values of α_{abs} in all spectral ranges. The work by Santos and Nogueira (2015) on this same site (campaign with filter measures in 2012–2013 in the Pantanal) states that during the wet season there is a predominance of biogenic aerosols, both in the coarse fraction and in the aerosol fine fraction. Here, measurements of the aerosol coarse fraction (PM10) were not performed, in this case, it can be stated that the fine fraction of biogenic aerosols has a spectral dependence of absorption less intense than what was observed during the dry season. More specific studies on biogenic aerosols need to be carried out in this region. Comparing the α_{abs} 580–880 nm obtained in the Pantanal with long-term measurements of α_{abs}, obtained by a linear adjustment for the ATTO site in the Central Amazon (Saturno et al., 2018), it was found that the values for the Pantanal are approximately 15% lower. Also, in comparison with the Central Amazon website (ZF2), Rizzo et al. (2013) found an average of 1.48 ± 1.12 for α_{abs} 450–700 nm for the wet season, while for the Pantanal α_{abs} 370–880 nm an average of 1.43 ± 0.49. These results indicate a strong similarity for the absorptive properties of aerosols between the Amazon and the Pantanal.

4. Conclusions

The principal contributors to the increase in scattering and absorption in the dry season were from local and regional fires (records from the Pantanal). The interannual comparison showed that an annual difference of approximately 70% in the burning records over the Pantanal can cause an increase of approximately 50% in the scattering and absorption coefficients. For ω_0 there was no statistical seasonal difference, however, the peculiar behavior, with medians below 0.8, at the begin-

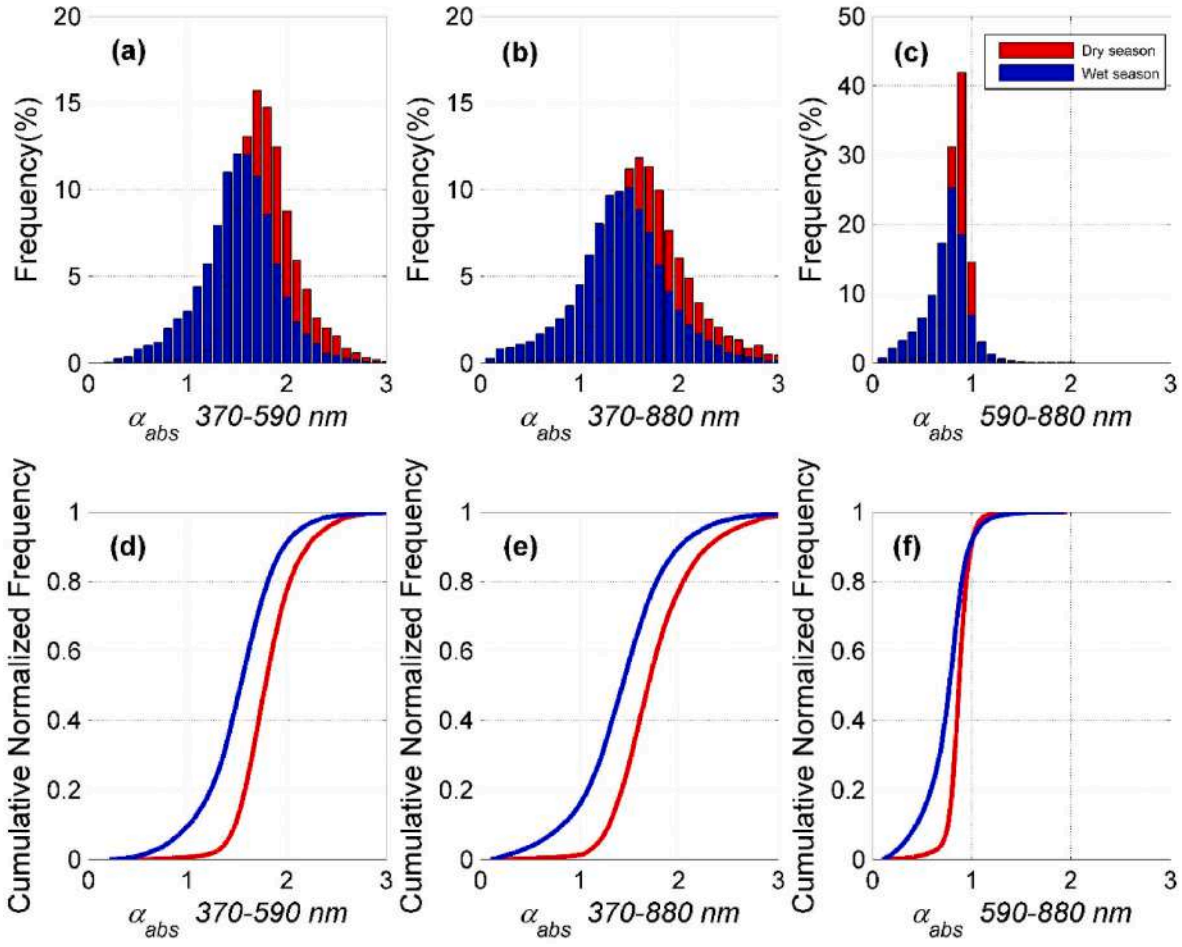

Fig. 7. Relative frequency for Angstrom exponents for the dry and wet seasons for the: a) α_{abs} 370–590 nm, b) α_{abs} 370–880 nm and c) α_{abs} 590–880 nm. Cumulative frequency normalized for Angstrom exponents for the dry and wet seasons for the: d) α_{abs} 370–590 nm, e) α_{abs} 370–880 nm and f) α_{abs} 590–880 nm.

ning of the peaks in local fires, indicates the strong absorption by the recent aerosols emission due to burning. As the dry season progresses, the values ω_0 rise again, due to a mixture of recent and aged emissions. In comparison, the values obtained for σ_{abs}, σ_{scat} and ω_0, during the wet season, within the limits of uncertainty, are practically equal to the values obtained for the central Amazon.

Mean BC concentrations obtained for the dry and wet seasons, were 0.75 ± 0.83 and 0.12 ± 0.09 μgm^{-3}, respectively. Compared to sites in the Amazon, these values were found to be approximately 70% lower than the concentrations found for Porto Velho, a site directly influenced by biomass burning in the Amazon. However, for the ZF2 site (central Amazon) the Pantanal concentrations were higher by 55% and 15%, for the dry and wet seasons, respectively. These findings support our conclusions that during the wet season the atmosphere of the Pantanal is as clean as that of central Amazonia in terms of BC concentration, and likewise, during the dry season it is more impacted, however, less than the regions directly affected by the Amazonian burning.

As expected, absorption coefficients decreased by a factor of 10 from dry to wet seasons at all wavelengths. This behavior was similar to results found for the central Amazon, a specific consequence of burning emissions during the dry season. The mean values of α_{abs} for the dry season showed strong contributions of BrC in radiative absorption. On the other hand, for the wet season (background conditions), the results showed a lower absorption spectral dependence than for the dry season. The results of this work suggest that the contributions of biogenic emissions, free from biomass burning, are responsible for the reduction of α_{abs} values in all spectral ranges. Compared to the results presented here, a strong similarity was found between the absorptive properties of aerosols for the Pantanal and for the Central Amazon.

Author contributions

Artaxo, P., and Nogueira conceived the experiment and coordinated research grants. Palácios, R., Rothmund, L., Basso, J. and Morais, F. conducted the measurements. Palácios, R., Romera, K. analyzed the data and wrote the manuscript. Rizzo, L., Cirino, G., Adams, D., Imbiriba, B., Nassarden, D., Siqueira, A., Rodrigues, T., Curado, L. and Weber, A. contributed to the discussion of the results. All authors reviewed the manuscript.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.apr.2022.101413.

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