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Strong influence of Black Carbon on aerosol optical properties in Central Amazonia during the fire season

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Abstract. During the dry season, the Amazonian atmosphere is strongly impacted by fires, even in remote areas. However, there are still knowledge gaps regarding how each aerosol type affects the aerosol radiative forcing. This work characterizes the chemical composition of submicrometer aerosols and source apportionment of Organic Aerosols (OA) and Equivalent Black Carbon (eBC) to study their influence on light scattering and absorption at a remote site in central Amazonia during the dry season (August-December 2013). We applied Positive Matrix Factorization (PMF) and multi-linear regression models to estimate chemical-dependent mass scattering (MSE) and extinction (MEE) efficiencies. Mean PM1 aerosol mass loading was 6.3±3.3 μg m^{-3}, with 77% of organics, grouped into 3 factors: Biomass Burning OA (BBOA), Isoprene derived Epoxydiol-Secondary OA (IEPOX-SOA) and Oxygenated OA (OOA). The bulk scattering and absorption coefficients at 637 nm were 17±10 Mm^{-1} and 3±2 Mm^{-1}, yielding a single scattering albedo of 0.87±0.03. Although eBC represented only 6% of the PM1 mass loading, MSE was highest for the eBC (13.58-7.62 m^2 g^{-1} at 450-700 nm), followed by BBOA (7.96-3.10 m^2 g^{-1}) and ammonium sulfate (AS, 4.79-4.58 m^2 g^{-1}). MEE was dominated by eBC (30.8%), followed by the OOA (19.9%) and AS (17.6%). The dominance of eBC over light scattering, in addition to absorption, depicts a surprisingly high role of this important climate agent, indicating the need to further investigate the chemical processing and interaction between natural and anthropogenic aerosol sources over remote tropical forested areas.

1 Introduction

The Amazon is the largest hydrologic basin and contiguous tropical forest area of the planet, representing important carbon reserves, sources of freshwater, and housing a large biodiversity (Andreae et al., 2015; Artaxo et al., 2013; Davidson et al., 2012; Pöhlker et al., 2016). The strong coupling between climate and the biological functioning of the forest is a key factor in the maintenance of the Amazonian ecosystem (Martin et al., 2010b; Pohlker et al., 2012). The Amazonian atmosphere is

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Preprint. Discussion started: 7 November 2024

considered an important reactor, regulating its physical properties and chemical composition due to the high insolation and humidity (Andreae, 2001). Despite the high temperatures, precipitation rates, and insolation during most of the year, an annual cycle can be observed, with a wetter and less warm season and a drier and hotter season, whose length and period vary depending on the region of the Amazon (Marengo et al., 2001).

During the wet season, the Amazonian atmosphere represents one of the few continental regions with episodic atmospheric composition near pristine conditions (Andreae et al., 2015; Martin et al., 2010b; Pöhlker et al., 2018; Pöschl et al., 2010). Particle number concentration during the cleanest period is a few hundred particles cm^{-3}, very similar to remote oceanic regions (Andreae et al., 2015; Artaxo et al., 2013; Martin et al., 2010). However, during the dry season, forest fires are provoked in order to clear forest areas for agriculture, and also as part of pasture and cropland management (Aragão et al., 2016; Berenguer et al., 2021; Davidson et al., 2012). During these periods, forest fire emissions coupled with smaller rates of aerosol scavenging lead to particle number concentration increases by a factor of 10 in remote forest areas (Artaxo et al., 2013; Pöhlker et al., 2018). These stark seasonal differences in aerosol loading and composition have the potential to significantly modify the coupling biosphere-atmosphere (Zaveri et al., 2022), which is expected to be exacerbated in the future due to extreme climatic events in Amazonia (Flores et al., 2024).

Atmospheric aerosol particles influence climate through scattering and absorption of solar radiation (aerosol-radiation interactions, ARI) and by affecting cloud formation and lifetime (aerosol-cloud interactions, ACI) (Forster et al., 2021). However, the magnitude and the signal of global radiative forcing of aerosols still represent one of the largest uncertainties in global climate models (Kuhn et al., 2010; Rizzo et al., 2013; Szopa et al., 2023). Uncertainties on the radiative forcing of individual aerosol components are even higher (Myhre et al., 2013), with a direct impact on the accuracy of future climatic scenarios (Forster et al., 2021). Carbonaceous aerosols (i.e. organic and black carbon) dominate the Amazonian atmosphere (Artaxo et al., 2013). While Organic Aerosol (OA) originates from both primary emissions, as well as secondary formation from gaseous precursors (Martin et al., 2010), black carbon is mostly primarily emitted from incomplete combustion, and in remote areas of the Amazon it is associated with Amazonian or transatlantic forest fires (Artaxo et al., 2013; Holanda et al., 2020). The sign and magnitude of the ARI forcing are dependent on several parameters such as particle origin, size distribution, mixture and age, notably affecting the light-absorbing component of OA, termed brown carbon (Laskin et al., 2015; Saturno et al., 2018b).

The secondary component of OA (SOA) in the Amazon has been shown to be a major component, notably during the wet season (Chen et al., 2015; Shrivastava et al., 2019). Isoprene (2-methyl-1,3-butadiene, C_5H_8) is the most abundant VOC emitted globally, mostly located in tropical forests such as the Amazon (Marais et al., 2016; Yáñez-Serrano et al., 2015). The formation of isoprene-derived Secondary Organic Aerosol (SOA) is a sequence of complex reactions and depends on different factors, such low concentrations of NO (a typical byproduct of fossil fuel combustion), and pre-existing aerosol particles where isoprene can condense on (Brito et al., 2018; Caravan et al., 2024; Marais et al., 2016; Nah et al., 2019). One of the dominating isoprene SOA pathways in the Amazon is through the OH attack, leading to hydroperoxy radicals and subsequently via the HO_2 pathway (Shrivastava et al., 2019; Wennberg et al., 2018). This pathway can lead to different low-volatility products

https://doi.org/10.5194/egusphere-2024-3339
Preprint. Discussion started: 7 November 2024

generally termed IEPOX-SOA (Isoprene EPOXydiols-Secondary Organic Aerosol) (Allan et al., 2014; Hu et al., 2015; Surratt et al., 2010). Isoprene oxidation product mixing ratios were previously shown to be higher during the dry season above the forest canopy, likely due to the higher insolation and temperature during this period, which favors the oxidative capacity of the atmosphere and leaf emission potential (Yáñez-Serrano et al., 2015). IEPOX-SOA mass concentrations have been shown to be significantly reduced during polluted conditions, associated with suppression due to urban NO emissions (de Sá et al., 2017). Alternatively, a study in polluted urban plumes in West Africa found that SO_4 increase plays a larger role in enhancing IEPOX-SOA loadings than NO in suppressing it (Brito et al., 2018). Secondary oxidized aerosol particles have been demonstrated to be more efficient at scattering radiation than primary particles (Kleinman et al., 2020; Paredes-Miranda et al., 2009; Reid et al., 2005; Smith et al., 2020).

Chemical composition, processes, and sources of atmospheric aerosol particles in the Amazon have been widely studied during both the wet and dry seasons, in sites representing pristine conditions (Andreae et al., 2015; Chen et al., 2015; Martin et al., 2010a) as well as strongly impacted by fires and urban pollution (Brito et al., 2014; Ponczek et al., 2021; de Sá et al., 2018; Zaveri et al., 2022). Physical properties of radiation absorption and scattering were described (Artaxo et al., 2013; Nascimento et al., 2021; Palácios et al., 2020; Rizzo et al., 2013; de Sá et al., 2019; Sena et al., 2013). However, the intrinsic optical properties of each aerosol species are still rare (Velazquez-Garcia et al., 2023), notably associated with OA origins (Ponczek et al., 2021). Our study details the chemical properties of submicrometer aerosol particles in a forest site in central Amazonia during the dry season and their influence on radiation scattering and absorption. We applied positive matrix factorization (PMF) to the organic fraction, associated mass extinction, absorption, and scattering efficiencies to different aerosol components via multi-linear regression to improve our comprehension of their intrinsic properties, as well as estimate their role on aerosol-radiation interaction in Central Amazonia during the dry season.

2 Material and Methods

2.1 Sampling site

The measurements site is located 60 km northwest of the city of Manaus, Brazil, in the Cuieiras biological reserve (2°35’39.24’’S, 60°12’33.42’’W), and referred to in this study as T0z (Martin et al., 2015; Whitehead et al., 2016). The vegetation is characterized as *terra firme* (upland forest, not impacted by seasonally flooded), and the canopy is between 30 m and 35 m high (Martin et al., 2010). As a result of steady northeasterly-easterly winds (Andreae et al., 2015; Araújo et al., 2002), only rarely the site is impacted by Manaus emissions (Chen et al., 2015). The seasonality at the region of this site in central Amazonia has been previously defined as the wet season from 1 December – 14 June, and the dry season from 15 June – 30 November (Andreae et al., 2015). During the wet season, air masses reaching the site pass over more than 1,500 km of undisturbed forest (Andreae et al., 2015; Pöschl et al., 2010). However, during the dry season, regional biomass burning pollution can be detected at the site (Artaxo et al., 2013; Rizzo et al., 2013), as well as aerosol plumes advected from African wildfires (Holanda et al., 2023). Our observations comprise from 1 August until 10 December 2013, sampling the atmosphere

https://doi.org/10.5194/egusphere-2024-3339
Preprint. Discussion started: 7 November 2024

at 38.8 meters above ground level. The instrumentation was located inside an air-conditioned container at the base of the tower. A cyclone (50 % cut-off at 10 µm) was used at the entrance of the inlet. An automatic diffusion dryer (Tuch et al., 2009) kept the relative humidity of the sampled air between 20% and 50%. Lodging for scientists/staff and a diesel generator were located 330m and 720m downwind (west) from the tower, respectively. The measurement tower has been shown to be practically unaffected by the generator (Whitehead et al., 2016). The year of this study (2013) was characterized by a historical minimum of fire detection over the last 20 years (Figure 1, (F. G. Assis et al., 2019)), providing an interesting outlook to assess the best scenarios for a dry season in recent times, and thus evaluate atmospheric composition within targets and goals for the Amazon forest preservation. The observation period here has been considered to fit entirely within dry-season atmospheric conditions. The previous transitional (wet-dry) period occurred in June-July (Whitehead et al., 2016), and the subsequent (dry-wet) soon after the end of our measurements.

https://doi.org/10.5194/egusphere-2024-3339
Preprint. Discussion started: 7 November 2024

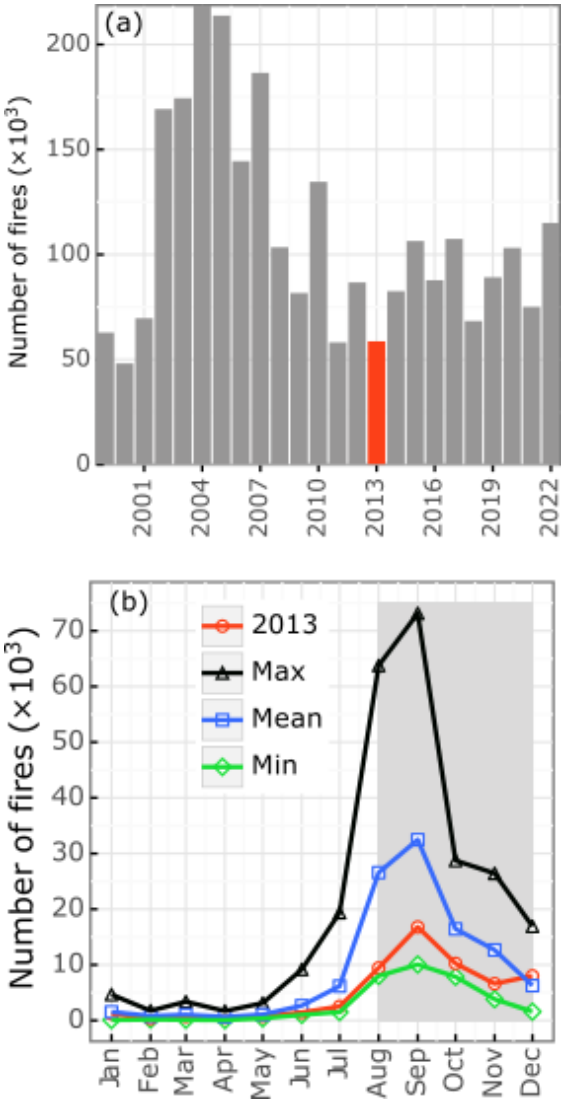

Figure 1: a) Number of fires in the Brazilian Amazon forest from 1999 to 2022, showing how 2013 (marked in red) is the lowest in the past 20 years in terms of total number of fires, and b) mean (blue), maximum (black), minimum (green), and for 2013 (red) monthly fires between 1999-2022 in the Amazon Basin. The gray area in b) marks the period of measurements in our study (01/Aug – 10/Dec) (Instituto Nacional de Pesquisas Espaciais, 2024)**.**

2.2 Instrumentation

Non-refractory submicrometer aerosol composition was measured using a quadrupole Aerosol Chemical Speciation Monitor (ACSM, Aerodyne Research Inc) (Ng et al., 2011), which is a compact version of the Aerosol Mass Spectrometer (AMS). Instrument calibration consisted of injecting monodispersed (300 nm) aerosol particles of ammonium nitrate (AN) and

https://doi.org/10.5194/egusphere-2024-3339
Preprint. Discussion started: 7 November 2024

ammonium sulfate (AS). Aerosol particles were generated using an atomizer and subsequently dried, and size selected using a Differential Mobility Analyzer. A collection efficiency of 0.5 has been adopted (Middlebrook et al., 2012), yielding a very good agreement in particle mass considering measurements from different collocated instruments (S1). This method was successfully used in previous studies (Brito et al., 2014; Sun et al., 2010), and the value of 0.5 agrees with other studies in the Amazon during the dry season (de Sá et al., 2019) and during the transition from wet to dry season (Ponczek et al., 2021). The measured ammonium (NH_4) mass concentrations were close to, or often lower than the detection limit of 0.28 μm^{-3} (Pöhlker et al., 2018; Whitehead et al., 2016), and were therefore calculated based on nitrate (NO_3) and sulfate (SO_4) molar masses and their mass concentrations, assuming neutralization as in Equation 1:

$$NH_{4,\ predicted} = 18 \times \left(\frac{SO_4}{96} \times 2 + \frac{NO_3}{62}\right) \quad (1)$$

Furthermore, the SO_4 and NO_3 ions were used to estimate AS and AN for the chemical-dependent optical properties analyses (Section 3.3), assumed here to be their most abundant form given the very low NH_4 levels:

$$AS = 132 \times \frac{SO_4}{96} \quad (2)$$

$$AN = 80 \times \frac{NO_3}{62} \quad (3)$$

Size-resolved particle number size distribution from 10 to 450 nm was measured with a Scanning Mobility Particle Sizer (SMPS, model 3081, TSI Inc.) coupled to a Condensation Particle Counter (CPC, model 3772, TSI Inc.) to provide equivalent mobility particle diameter for singly charged particles (D_{pg}, (Wiedensohler et al., 2012). Aerosol light scattering coefficient (σ_s) at 450 nm, 550 nm, and 700 nm (Anderson and Ogren, 1998) was measured using a Nephelometer (model 3563, TSI Inc.). Calibration was performed using CO_2 as the high-span gas and filtered air as the low-span gas. The averaging time applied was 60 minutes, and therefore the detection limits, defined as a signal-to-noise ratio of 2, for scattering coefficients are 0.08, 0.03, and 0.05 Mm^{-1} for 450, 550, and 700 nm, respectively (Anderson and Ogren, 1998). Since a PM10 inlet was used, the "no-cut" factors were used for the truncation corrections (Anderson and Ogren, 1998). Scattering coefficients at 637 nm were calculated from interpolation, assuming a power-law spectral dependency. A Multi Angle Absorption Photometer (MAAP, model 5012, Thermo Electron Group, Waltham, USA) (Müller et al., 2011) measured aerosol light absorption coefficient (σ_a) at 637 nm and was used to estimate equivalent Black Carbon (eBC) concentration, assuming an absorption cross-section value of 6.6 $m^2\ g^{-1}$. Considering the conditions of the experiment, the MAAP detection limit for σ_a was of 0.13 Mm^{-1} (Petzold et al., 2005).

Episodes of possible contamination from the city of Manaus and from the diesel generator were removed by filtering the datapoints when either the wind direction was between 270-340° (from our local wind direction measurements) or when the

https://doi.org/10.5194/egusphere-2024-3339
Preprint. Discussion started: 7 November 2024

calculated backtrajectories from the Hysplit model (Draxler and Hess, 1998) passed over Manaus coordinates, as in (Whitehead et al., 2016) (Supplement S2).

2.3 Optical properties

Single Scattering Albedo (SSA, Equation 4) is a measure of the ratio of σ_s to the total radiation extinction coefficient ($\sigma_e = \sigma_s + \sigma_a$) by aerosol particles (Rizzo et al., 2013). Since the MAAP instrument only measures the σ_a at 637 nm wavelength, the σ_e was calculated only for this wavelength using σ_s at 637 nm from the nephelometer, estimated using the scattering Angstrom exponent (α_s, Equation 5).

$$SSA = \frac{\sigma_s}{\sigma_s + \sigma_a} \quad (4)$$

The α_s is a measure of the dependence of radiation scattering on the light wavelength (λ), and it is an indication of particle size (Rizzo et al., 2013; Saturno et al., 2018b; Schuster et al., 2006):

$$\ln \sigma_s = -\alpha_s \ln \lambda + \ln (constant) \quad (5)$$

After rain events and other moments when the atmosphere is very clean, both α_s for σ_e values are very low, and therefore the ratio between them (SSA, Equation 4) becomes unrealistically high. We therefore calculated SSA for α_s for $\sigma_e > 1$ Mm^{-1}.

2.4 Statistical Analyses

2.4.1 Positive Matrix Factorization (PMF)

The PMF (Positive Matrix Factorization) was used on the submicrometer non-refractory organic mass spectra in order to group m/z ratios with similar temporal variability, supporting the identification of sources and processes that formed and transformed atmospheric particles. A detailed description of the PMF can be found in (Paatero & Tapper, 1994; Ulbrich et al., 2009), and it is an established methodology for aerosol source apportionment (Zhang et al., 2011). The model can be represented by the following Equation. (6):

$$X_{(m \times n)} = \sum G_{(m \times p)} \cdot F_{(p \times n)} + \mathrm{E} \quad (6)$$

Where **X** is the input matrix of *n* (elements – m/z ratios) lines and *m* (number of samples) columns (Ulbrich et al., 2009). In this study, the **X** matrix had 2901 lines (1-hour averages for more than 4 months of measurements) and 70 columns (m/z ratios). The receptor model aims to determine the number of *p* factors, representing sources or processes, their chemical composition, and the relative contribution of each factor. **G** is a matrix in which columns are the time series of the factors. **F** is a matrix in which lines are the profiles of the factors (mass spectra). **E** represents the residuals, the part of the data that was

https://doi.org/10.5194/egusphere-2024-3339
Preprint. Discussion started: 7 November 2024

not modeled by any factor *p*. We used an IGOR™-based interface to apply the PMF analysis (Ulbrich et al., 2009). The PMF ions were normalized to the organics concentration.

2.4.2 Multilinear Regression (MLR)

We used a Multilinear Regression (MLR) model to estimate the contribution of each aerosol chemical component to scattering, absorption, and extinction coefficients, deriving the corresponding efficiencies (MSE, MAE, and MEE, respectively). The scattering (σ_s) and absorption (σ_a) coefficients (Mm^{-1}) measured with the nephelometer and the MAAP (respectively) were the dependent variables (response) and the species/factors were the independent (predictor) variables.
A generalization of the mass efficiency (ME) calculation is presented in Equation 7:

$$ME = \sum_i a_i x_i + r \quad (7)$$

Where ME can be MSE, MAE or MEE; *x* is the chemical species mass concentration; a_i is the efficiency of each component, and *r* are the residuals. We used NNLS (Non-Negative Least Squares) from Python package Scipy version 1.5.2 (Virtanen et al., 2020). To constrain the model to produce results with physical meaning, the coefficients a_i were constrained to be positive, as in (Velazquez-Garcia et al., 2023). Since eBC is assumed to be the only absorbing component measured in this study with the MAAP, a MLR could not be applied, and the MAE was considered to be equivalent to the cross-section value (6.6 m^{-2} g^{-1}, Section 2.2).

3 Results and Discussion

3.1 Aerosol chemical composition

The total submicrometer (PM1 = Organics + NO_3 + NH_4 + SO_4 + eBC) mean mass concentration during the observation period was 6.3±3.3 μg m^{-3} (Table 1, Figure 1a). This represents about half of what was measured during the dry season of the following year (2014, with much more fires, Figure 1) at a nearby site (ATTO tower, Amazon Tall Tower Observatory), with similar conditions (Central Amazonia, isolated from major biomass burning focus or Manaus urban plume) (10.5 μg m^{-3}, (de Sá et al., 2019)) and regions directly impacted by fires during the dry season (12.4-13.7 μg m^{-3} (Brito et al., 2014; Ponczek et al., 2021)). The PM1 aerosol composition was dominated by the organic fraction, representing a mass fraction of 77±5% (Table 1, Figure 2b). This is very similar to the wet season and wet to dry season transition in the same site (> 80% (Artaxo et al., 2013; Chen et al., 2015; Whitehead et al., 2016)), but high compared to other continental urban areas, such as across Europe (~30-50%, (Chen et al., 2022)) and lower than the strongly impacted by biomass burning region in Southwestern Amazonia (90%, (Brito et al., 2014). Sulfate is the main soluble inorganic component of the aerosol mass fraction in the Amazon, both during the wet and dry seasons (Fuzzi et al., 2007; Yamasoe et al., 2000). In our study, the mean SO_4 mass fraction was 9±3% (Table 1, Figure 2b), which is comparable to the ATTO site (Andreae et al., 2015). However, in Southwestern Amazonia, in areas

https://doi.org/10.5194/egusphere-2024-3339
Preprint. Discussion started: 7 November 2024

impacted by fresh biomass burning, the average SO_4 mass fraction was significantly lower (2-3% (Brito et al., 2014; Ponczek et al., 2021)).

The eBC mass fraction was 6±2% (Table 1, Figure 2b), which is half than the fraction observed in the wet season at the same site (11%), despite the much lower total submicrometer aerosol mass loading (Chen et al., 2015). Occasional urban pollution from Manaus, long-range transport from Africa, and potential artifacts combined with much lower overall aerosol loading are possible causes for this relatively higher eBC mass fraction in the wet season (Chen et al., 2015). In Southwestern Amazonia, during the transition from dry to wet season, the contribution of eBC to PM1 reached 15% (Ponczek et al., 2021). Nitrate had a minor contribution during our observations (3±1%), with concentration levels comparable to the ATTO site (Pöhlker et al., 2018), as well as Southwestern Amazonia (Brito et al., 2014; Ponczek et al., 2021)). The concentrations of organics and inorganics follow similar patterns during the measurement period (Figure 2a). This can be an indication that the total mass loading consists of well-mixed biomass burning and secondary aerosols, associated with large and regional-scale processes (Darbyshire et al., 2019).

https://doi.org/10.5194/egusphere-2024-3339
Preprint. Discussion started: 7 November 2024

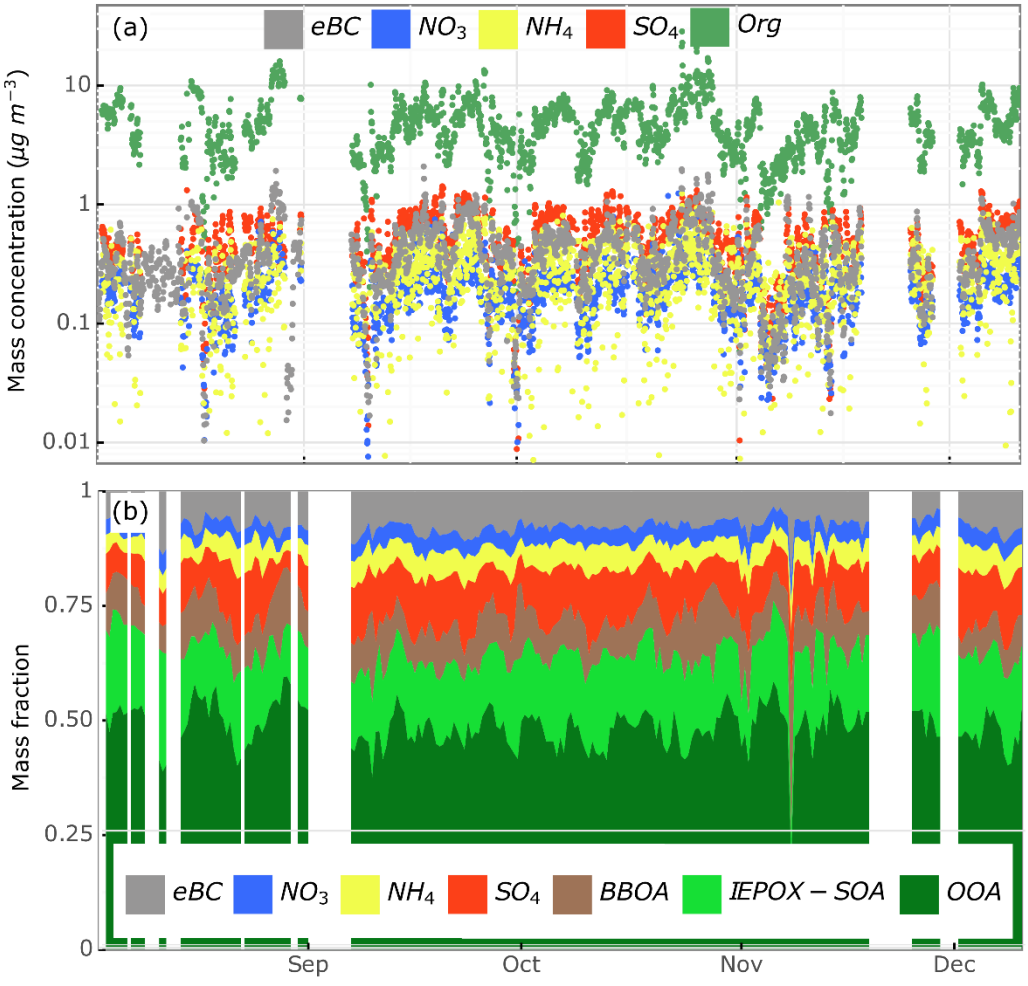

Figure 2: Non-refractory submicrometer aerosol species and eBC mass (a) concentrations and (b) fractions at T0z. In (a) the vertical axis is the logarithm scale to facilitate the visualization of different species. In (a), data is shown in the original time stamp of 1 hour, while in (b) it was averaged to 12 hours in order to facilitate visualization.

The PMF analysis yielded 4 factors, although 2 statistical factors were closely related to the Oxygenated Organic Aerosol (OOA) fraction, and the mass spectra, diurnal profile, and time series of these factors did not present enough differences to justify their separation (Supplements S3 and S4). Therefore, these 2 factors were manually summed in order to generate a 3 factors solution, which was different from the factors found in the 3 factors solution presented by the PMF. In (Ulbrich et al., 2009), the authors describe how one PMF resulting statistical factor can split into various other factors which, after added, represent the real factor. The recombination often considers similarities between the statistical factors in the mass spectra, diurnal profile and time series (Carbone et al., 2013). In our study, the identification of the factors was further confirmed with the correlation between the PMF statistical factors and the inorganic aerosols, with the eBC (Supplement S4) and diurnal

https://doi.org/10.5194/egusphere-2024-3339
Preprint. Discussion started: 7 November 2024

profile analyses. The 3 factors were identified as BBOA (Biomass Burning Organic Aerosol), OOA (Oxygenated Organic Aerosol) and IEPOX-SOA (Isoprene derived Epoxydiol–Secondary Organic Aerosol), and they represent together 99% of the measured submicrometer organic aerosol mass, with 1% of residuals. The correlation between BBOA and NO_3 and SO_4 is comparable with findings in Southwestern Amazonia (Brito et al., 2014).

Table 1 - Dry season (01/Aug – 10/Dec) mean mass concentration (μ m^{-3}), standard deviations, and percentile range (10-90, in parenthesis) of the species measured by the ACSM and MAAP (eBC).

	Mass concentration (μg m^{-3})	Mass fraction
Total PM1	6.3±3.3 (2.7-10.3)	100%
Organics	4.9±2.7 (2.1-7.9)	77±5%
BBOA	*0.6±0.6 (0.2-1.1)*	9±5%
IEPOX-SOA	1.0±*0.5 (0.4-1.7)*	*17*±5%
OOA	3.2±*1.5 (1.3-5.5)*	*51*±6%
NO_3	0.2±0.1 (0.1-0.3)	3±1%
NH_4	0.3±0.1 (0.0-0.5)	4±1%
SO_4	0.5±0.3 (0.2-0.9)	9±3%
eBC	0.4±0.3 (0.1-0.7)	6±2%

The largest contribution PMF-derived statistical factor in our study was OOA, which contributed to 51±6% of the PM1 (Table 1), and 65% of the organic mass. It presented the largest m/z 44 fraction (Figure 3), and therefore, this is the factor with the highest estimated O:C ratio (Chen et al., 2015). The high oxidation level indicates highly aged particles and may lose some of their original chemical signatures, in terms of elementary ratios, during the aging process (Jimenez et al., 2009). The m/z 44 is formed mainly by the fragment CO_2^+, typical of the thermal decarboxylation of the organic acids groups (Alfarra et al., 2004). The more aged the aerosols, the more chemically similar they become, which makes the task of separating them into different factors with distinctive characteristics very difficult. Therefore, the OOA factor probably groups aerosol particles from different sources, and their common characteristic is that they are probably originating relatively distant from the sampling site. This factor is a major component of basin-wide haze observed during biomass burning season, associated with the transport from eastern Amazonia into remote central regions at the center of the basin (Darbyshire et al., 2019).

https://doi.org/10.5194/egusphere-2024-3339
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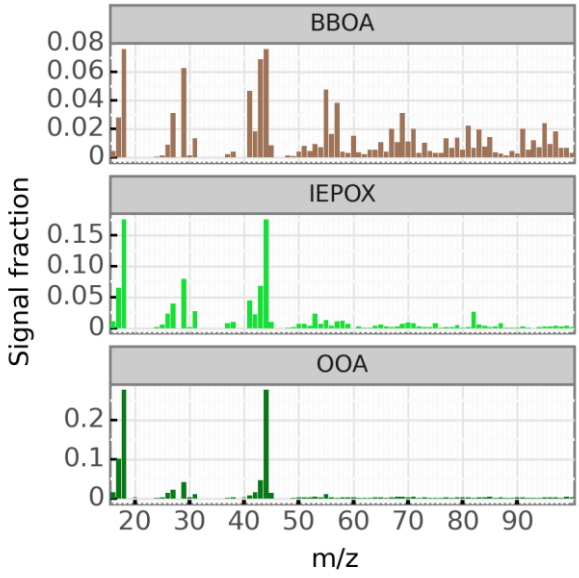

Figure 3: PMF Mass spectra composition of each statistical factor and its relative contribution to the total submicrometer organic aerosol mass. It is possible to observe typical tracer ions such as m/z 60 and m/z 73 that characterize the BBOA factor, and also the m/z 82 in the IEPOX-SOA factor.

The production of IEPOX-SOA generally leads to the production of markers in the atmosphere, such as the 2-methylthetrol and the 3-methylfuran (m/z 82, $C_5H_6O^+$). These markers may not originally exist in the IEPOX-SOA molecule due to their extremely high volatility, but they can be formed during the decomposition of some IEPOX-SOA species, such as 3-methyltetrahydrofuran-3,4-diols (3-MeTHF-3,4-diols) (Lin et al., 2012). The organic fraction in the m/z 82 is therefore important for the identification of the IEPOX-SOA factor, despite its low contribution to the submicrometer organic aerosol mass fraction (usually below 4%). Beyond that, most of the other m/z are common to other factors, making the m/z 82 distinctive of the IEPOX-SOA, which can also be identified by the m/z 53 ($C_4H_5^+$) and m/z 75 ($C_3H_7O_2^+$) (Allan et al., 2014; Lin et al., 2012; Xu et al., 2015a). The lifetime of IEPOX-SOA in the boundary layer is estimated to be about 2 weeks (Hu et al., 2016). The IEPOX-SOA mean mass concentration in our study was 1.0±0.5 μg m^{-3} (Table 1, Figure 4). Previous studies reported 0.26 μg m^{-3} during the wet season (Chen et al., 2015), while downwind of Manaus it was around 0.5 μg m^{-3} during background conditions, and 0.1 μg m^{-3} during polluted conditions (de Sá et al., 2017). It is important to note that at T0z, while the organic particle loading typically increases by an order of magnitude from the wet to the dry season (Artaxo et al., 2013), we estimated that IEPOX-SOA increases about a factor ~3. The relative contribution of IEPOX-SOA to organics during the wet season was estimated to be 34%, in contrast to the 17% found in this study (Table 1), which is likely the result of a complex balance between increased isoprene emissions (Yáñez-Serrano et al., 2015), sulfate abundance and increased pollution levels (including NO_x from forest fires, and biomass-burning related aerosol particles). The relative contribution of IEPOX-SOA to the total PM1 mass was relatively constant during the whole measurement period (Figure 2b), as well as most of the other

https://doi.org/10.5194/egusphere-2024-3339
Preprint. Discussion started: 7 November 2024

species (with the exception of some episodes). This indicates that an atmospheric dynamics of rain/dilution controlling the chemical composition could be more important than the influence of local sources of particles, confirming the regional haze hypothesis raised by (Darbyshire et al., 2019).

The correlation (Pearson coefficient = 0.7) observed between the IEPOX-SOA factor and AS (Supplement S4.1) is similar to the correlation measured in regions affected by urban pollution in the Amazon, Africa and USA (R^2 = 0.37-0.48 (Brito et al., 2018; Budisulistiorini et al., 2013; de Sá et al., 2017)). Sulfate is the main aqueous phase particle in which isoprene products use to condense on (Budisulistiorini et al., 2013; Kroll et al., 2006; Lin et al., 2012; Marais et al., 2016; Surratt et al., 2010; Xu et al., 2015b), and therefore a positive and moderate-high correlation is expected. eBC presents a similar correlation with IEPOX-SOA as AS (Supplement S4), but the correlation is even higher with the other PMF statistical factors, especially OOA (Pearson coefficient = 0.85, Supplement S4). Since OOA corresponds to more than half of PM1 (Table 1), this high correlation indicates that most of the submicrometer aerosols measured during the dry season in Central Amazonia are, in general, influenced by biomass burning emissions, since eBC is an important combustion tracer. However, the fact that the eBC correlation is higher with the OOA factor than with BBOA (which constitutes only 9% of PM1, Table 1, Supplement S4) indicates that long-range transport of aged and internally well mixed biomass burning plumes plays a more important role than nearby fire sources (Darbyshire et al., 2019).

The BBOA factor can be identified by the presence of the m/z 60 and m/z 73 (Figure 3), which are dominated by the $C_2H_4O_2^+$ and the $C_3H_5O_2^+$ fragments. These fragments are originated from levoglucosan and other similar anhydro-sugars (such as manosan and galactosan). Levoglucosan (1,6-*â*-D-anhydroglucopyranose, $C_6H_{10}O_5$) is known as a biomarker of biomass burning emissions due to its production from the pyrolysis of carbohydrates as cellulose (Alfarra et al., 2007; Artaxo et al., 2013; Chen et al., 2009; Lee et al., 2010). The signal fraction of m/z 60 for the BBOA factor in our study was 1.5%, which is 5 times higher than the 0.3% threshold typically used as an appropriate background fraction for biomass burning (Cubison et al., 2011). OOA presented a m/z 60 signal fraction of 0.2%, while IEPOX-SOA presented a negligible signal.

The BBOA diurnal profile is different from that of the other PMF factors (Figure 4). While the OOA and IEPOX-SOA mass concentrations increase during the daytime due to photochemical oxidation processes, the BBOA mass concentration is fairly constant (Figure 4). Since the BBOA is a biomass-burning indicator, it is composed of mostly primary particles, so its concentration does not depend on photochemical activity. This pattern is different than the stark decrease in mass concentrations of fresh biomass burning particles during daytime, observed in southwestern Amazonia (Brito et al., 2014), where there were constant local sources of fires and the diurnal cycle was mostly determined by the boundary layer increasing and diluting the particles in a bigger area during daytime (Andreae et al., 2015). The lack of a clear diurnal pattern in our study for BBOA seems to confirm the regional origin of the aerosol particles, likely transported from distant biomass burning sources in the eastern parts of the basin, and long-range transport with complex interactions between residual and nocturnal layers (Darbyshire et al., 2019). An additional confirmation of the long-range transport is the relatively flat pattern of the eBC diel cycle, although there is a small but noticeable increase in the eBC diurnal mass concentration during daytime (Figure 4), which may indicate some lensing effect due to the increase in the particle coating (Denjean et al., 2020). While the diel cycle of the

https://doi.org/10.5194/egusphere-2024-3339
Preprint. Discussion started: 7 November 2024

NH_4 and NO_3 show practically no variation, the SO_4 indicates the influence of photochemical processes (Figure 4) or atmospheric transport. The higher boundary layer in the afternoon (Fisch et al., 2004) may favor the downward transport of long-distance particles (Darbyshire et al., 2019). An additional possible explanation for this observed increase in SO_4 during the afternoon is biogenic sources of SO_4 precursors.

As the OOA and the IEPOX-SOA factors represent together around 68% of the total mass fractions of the submicron particles during our study, and conversely, eBC and BBOA represent only 15%, the importance of the atmospheric photochemical activity in Central Amazonia becomes evident. Well-preserved parts of the Amazon are strongly affected by the regional transport of well-processed biomass burning plumes, overwhelming the local biogenic processes that usually modulate the diurnal behavior of secondary aerosol development (Artaxo et al., 2013; Darbyshire et al., 2019).

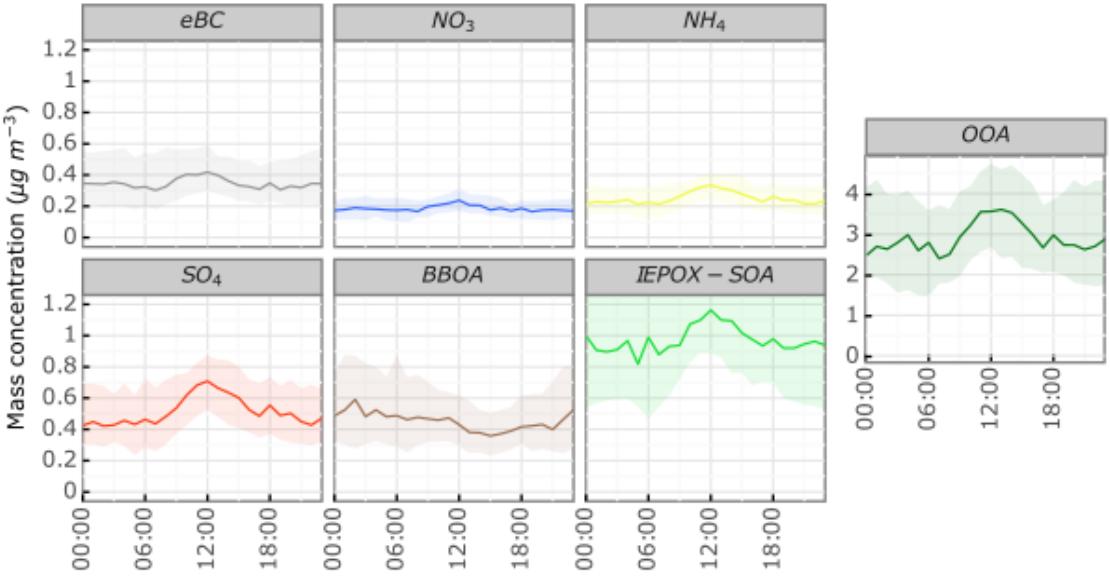

Figure 4: Diurnal profile (local time) of the PMF derived statistical factors, the inorganic chemical species and eBC mass concentrations for the whole period of measurements (1 August – 10 December, 2013). The lines represent mean values, and the shaded areas represent the standard deviations. The OOA factor, shown separately, has a different vertical scale.

3.2 Physical properties

The mean scattering coefficient at 637 nm in our study was 17±10 Mm^{-1} (Table 2), which is similar to the values reported for the same site and at the ATTO site during the dry season in previous years (Rizzo et al., 2013) and lower than observations close to biomass burning sources (32-80 Mm^{-1} (Artaxo et al., 2013; Ponczek et al., 2021)). In the dry season, fine mode particles predominate and are more efficient at scattering radiation than coarse mode dominated biogenic particles in the wet season (Rizzo et al., 2013). The absorption coefficient mean value was 3±2 Mm^{-1} (Table 2, Figure 5, Section 2.2).

https://doi.org/10.5194/egusphere-2024-3339
Preprint. Discussion started: 7 November 2024

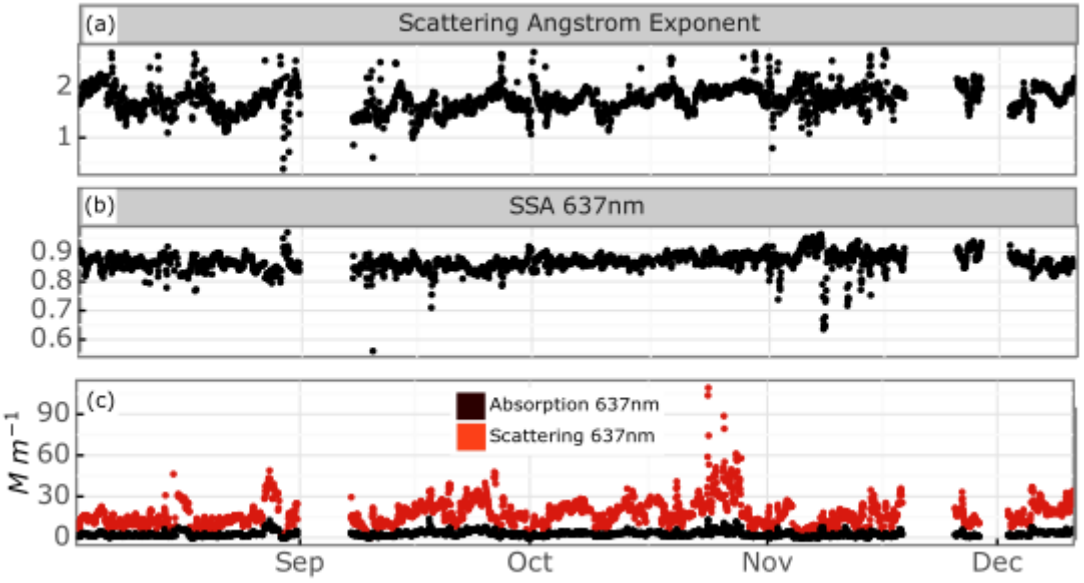

Figure 5: Time series of: a) the Scattering Angstrom Exponent (unitless), b) Single Scattering Albedo (SSA) at 637 nm (unitless), and c) absorption and scattering coefficients at 637 nm (Mm^{-1}).

The mean SSA observed in our study (0.87±0.03, Table 2) was very similar to the SSA reported for the nearby ATTO site (Saturno et al., 2018b). In previous years (2009-2012) at the same site as in our study, SSA varied from 0.84 to 0.91 from the wet to the dry season respectively. The dominance of organics and the relatively high SO_4 fraction in our study (9%, Table 1) are probably important factors for the high SSA (Artaxo et al., 2013; Rizzo et al., 2013). SSA was lower (0.77±0.08 at 637 nm) during the transition from dry to wet season in a site highly impacted by fires in southwestern Amazonia (Ponczek et al., 2021).

The mean value for the scattering Angstrom exponent in our study was 1.76±0.26 (Table 2), which is very similar to the 1.70±0.41 and the 1.71±0.24 measured in the dry seasons of previous years at the same site and at the nearby ATTO station (Rizzo et al., 2013; Saturno et al., 2018b), and the 1.65±0.37 measured during the dry season at a site more impacted by forest fires (Ponczek et al., 2021). However, it was higher than the 1.48±1.12 (although within the high variability range) and the 1.29±0.50 measured in wet seasons of previous years at the same station and the ATTO site (Rizzo et al., 2013; Saturno et al., 2018b). Higher scattering Angstrom exponent values are usually related to a greater proportion of fine mode particles in the aerosol population (Andreae et al., 2015), and in our case, it is probably related to the occurrence of fresh biomass burning particles.

Table 2 – Optical properties mean and standard deviation (in parentheses) for the whole study period for different wavelengths.

https://doi.org/10.5194/egusphere-2024-3339
Preprint. Discussion started: 7 November 2024

Optical property	Wavelength (nm)	Mean
Scattering coefficient (Mm^{-1})	450	32±19
Scattering coefficient (Mm^{-1})	550	22±13
Scattering coefficient (Mm^{-1})	637	17±10
Scattering coefficient (Mm^{-1})	700	14±8
Absorption coefficient (Mm^{-1})	637	3±2
Single Scattering Albedo (Mm^{-1})	637	0.87±0.03
Scattering Angstrom Exponent		1.76±0.26

3.3 Chemical-dependent optical properties

We applied the multiple linear regression (Section 2.4.2) to our dataset and the resulting coefficients successfully predicted the observed scattering (Figure 6), confirming the validation of this methodology to estimate the specific contribution of each chemical group to the optical properties. All the coefficients of the multilinear regression of all the wavelengths were statistically significant ($p < 0.001$) for both MSE and MEE. The MSE of the PMF factors decreased as a function of wavelength (Figure 7), in agreement with a previous study (Ponczek et al., 2021). Regarding the inorganic species, this decrease was also observed in the AN, but absent in the AS, which showed no wavelength dependency (Figure 7). The highest MSE values were attributed to eBC (Table 3, Figure 7a), followed by the BBOA. The MSE values of the eBC, BBOA and OOA components calculated in our study at 637 nm were *circa* 180%, 67% and 43% respectively compared to (Ponczek et al., 2021). MSE of AS in our study was between 4.58-4.79 $m^2\ g^{-1}$ (considering wavelengths from 450-700 nm, Table 4), which is almost double of the 2.5±0.6 $m^2\ g^{-1}$ fine mode average of 93 observations of a variety of regions (urban, remote, rural continental, ocean/marine) and seasons, and using different methods, mostly at 550 nm (Hand and Malm, 2007). Despite the small variability, lower MSE was reported for drier and cleaner environments, and higher MSE was found in more polluted regions and larger particle sizes (Hand and Malm, 2007), which may partially explain the higher values found in our study – in a wet climate and during the forest fires season when the atmosphere can resemble that of highly polluted regions (Artaxo et al., 2013). A recent study in an area highly impacted by urban pollution in France reported MSE for AS ranging from 4.8-7.1 $m^2\ g^{-1}$ (450-635 nm) (Velazquez-Garcia et al., 2023), resembling more similar results to our study for the lower wavelengths. Concerning the contribution of AN to the PM1 mass concentration, we tested the MLR removing AN, and the results were comparable, especially for eBC (Supplement Table S5.1). We also tested the robustness of the method by running 100 times MLR on random 50% of the data, yielding similar results (Supplement Table S5.2).

Aerosol particles which typically absorb radiation are also known to have significant scattering efficiencies, highly dependent on their sizes (Bond and Bergstrom, 2006). Biomass burning aerosols have been previously associated with high scattering efficiencies (Hand and Malm, 2007; Malm et al., 2005). However, no clear particle size dependency was observed for the

https://doi.org/10.5194/egusphere-2024-3339
Preprint. Discussion started: 7 November 2024

radiation scattering in our study (Figure 6). Our result of a pronounced MSE of the eBC represents an opposite trend than the observed in the transition from the dry to the wet season at a site more impacted by the fires, where the MSE of BBOA was higher than eBC's (Ponczek et al., 2021).

When the mass concentration is considered, the relative contribution of eBC to the scattering in all the measured wavelengths is about 20-25% or the total scattering (Figure 7b), comparable to Southwestern Amazonia (Ponczek et al., 2021), despite our site having a significantly lower eBC concentration. The contribution of AS and AN to MSE was from 20% to 30% with increasing wavelength (Figure 7b), less than half of that in urban sites in Europe (e.g. 67% in Northern France, (Velazquez-Garcia et al., 2023), but about twice as high as during an extreme pollution haze episode in Beijing (Wang et al., 2015).

When considering the total light extinction (scattering + absorption), the relative contribution of eBC reaches about 30% (Figure 8), which is comparable to the work in highly urbanized region in Europe (Velazquez-Garcia et al., 2023), however significantly lower than the 76±20% observed in urban pollution in China (Yu et al., 2010). By using the MSE and MEE ratios, we calculated specific SSA for the eBC, obtaining a value of 0.57. This means that 57% of the light extinction provoked by the eBC is scattered rather than absorbed, which is higher than the eBC specific SSA of 0.46 based on previous studies in more polluted conditions (Ponczek et al., 2021; Velazquez-Garcia et al., 2023). SSA of eBC has been described as typically ranging from 0.3 to 0.4, with higher values associated to heavy coating (Luo et al., 2020). Evidence of coating increasing elemental carbon scattering efficiency has been found in the past (He et al., 2015; Yu et al., 2010), however other studies found that thick coating in eBC lowered scattering coefficients (Darbyshire et al., 2019).

A previous study found that eBC absorption cross-section for the Amazon was 12.3 m^2 g^{-1} (Saturno et al., 2018b), and we tested our dataset applying this value. The result is that eBC mass concentrations would become half of what they are reported, with no change in σ_a, SSA, but MSE would double, while eBC contribution to MEE (Figure 8) would remain unchanged. Due to some methodology differences between our study and (Saturno et al., 2018a) (they measured refractory Black Carbon using a single-particle soot photometer SP2, with a higher cut-off, possibly leading to a sub-estimation of the mass), and the fact that applying the absorption cross-section value they found would make MSE of eBC be an order of magnitude higher than the others (Supplement Figure S6), we opted to remain with the more stablished value of 6.6 m^2 g^{-1}.

The OOA factor presented a relatively high relative contribution to MSE and MEE (Figures 8b and 9) due to its high mass relative contribution to the total PM1 mass (Table 1), although its MSE is relatively low (Figure 7a). Our results show an increase in AS relative fraction of contribution to MSE with increasing wavelength (from 10% to 20%, Figure 7b), while OOA decreased (from 30% to 20%, Figure 7b). The relative contributions to MSE of the OOA and BBOA were about twice as high in a site more directly impacted by fires during the dry season than in our study (Ponczek et al., 2021).

Table 3 – MEE, MAE and MSE for different wavelengths and aerosol components with standard errors. All the coefficients were statistically significant for all the wavelengths for MSE and MEE (p < 0.001).

	MSE (m^2 g^{-1})				MAE	MEE
Wavelength (nm)	450	550	637	700	637	637

https://doi.org/10.5194/egusphere-2024-3339
Preprint. Discussion started: 7 November 2024

eBC	13.58±1.08	10.67±0.70	8.68±0.52	7.62±0.44	6.6	15.28±0.52
BBOA	7.96±0.33	5.33±0.21	3.83±0.16	3.10±0.13		
IEPOX-SOA	5.61±0.41	3.84±0.27	2.87±0.20	2.37±0.17		
OOA	3.58±0.15	1.94±0.10	1.24±0.07	0.90±0.06		
AS	4.79±0.62	4.79±0.41	4.77±0.30	4.58±0.25		
AN	7.07±1.41	5.17±0.92	4.41±0.68	3.93±0.58		

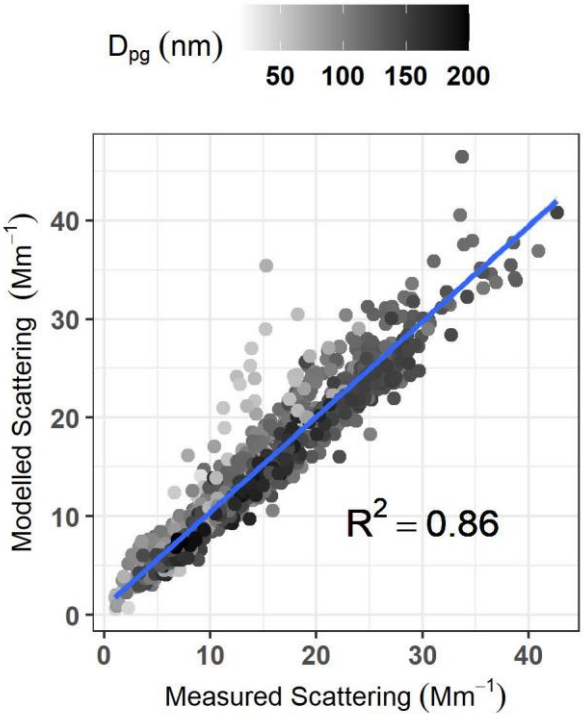

Figure 6: Measured radiation scattering at 637 nm vs modeled scattering (MSE). The gray scale corresponds to the equivalent mobility particle diameter for singly charged particles ($D_{pg,}$ section 2.2). The blue line indicates a linear correlation.

https://doi.org/10.5194/egusphere-2024-3339
Preprint. Discussion started: 7 November 2024

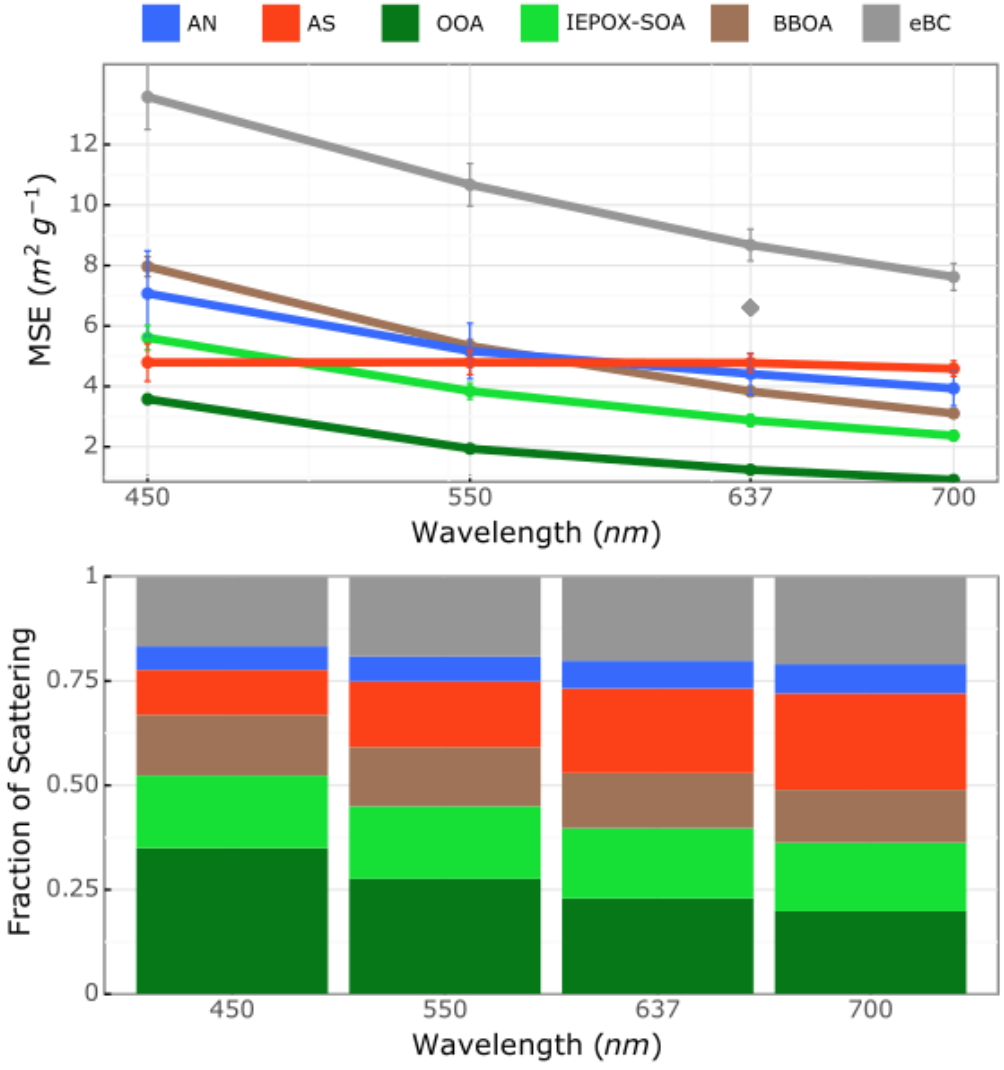

Figure 7: a) Mass scattering efficiencies (MSE) for each chemical component (color lines) at each wavelength, where the dots are the coefficients of the multi-linear regression, the bars are the standard errors, and the diamond shape is the value of absorption efficiency of the eBC at 637 nm; and b) the fraction of scattering of each component at each wavelength, considering its mass fraction to the total mass of submicrometer particles.

https://doi.org/10.5194/egusphere-2024-3339
Preprint. Discussion started: 7 November 2024

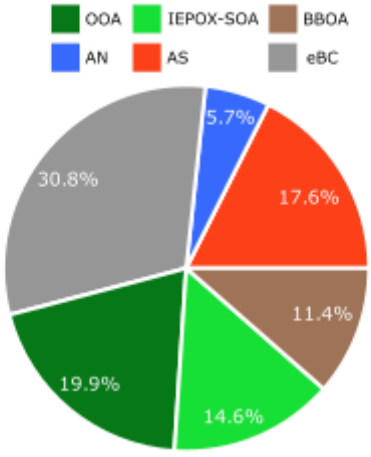

Figure 8: The relative contribution (%) of extinction coefficient (scattering + absorption) of each component considering its mass fraction to the total mass of submicrometer particles.

4 Conclusions

The results have shown that the submicrometer particle mass concentration during the dry season (6.3±3.3 μg m^{-3}), which is about an order of magnitude higher than typically observed at this site during the wet season, is highly dominated by the organic fraction (77±5%). The organic aerosols were separated into 3 PMF statistical factors, identified as BBOA, OOA, and IEPOX-SOA. The OOA, associated with highly processed and oxidized particles is the dominant factor (51±6% of the PM_1), followed by IEPOX-SOA (isoprene SOA following a low-NO route,17±5%), while the factor more directly associated with fresh biomass burning emissions (BBOA) represents 9±5% of PM1.

The mean radiation scattering coefficient at 637 nm was 17±10 Mm^{-1}, and the mean absorption coefficient was 3±2 Mm^{-1}, which are lower than the respective coefficients measured at sites much more impacted by the proximity to fresh forest fires but exceed those observed during the wet season. The SSA of 0.87±0.03 was elevated compared to values previously described in the wet season, but generally in good agreement with SSA values of previous dry seasons. The mean scattering Angstrom exponent was 1.76±0.26, surpassing the values previously measured during the wet season, likely due to the greater relative proportion of smaller particles in the aerosol population measured in our study, compared to the primary biogenic aerosols, Saharan dust and sea salt typical of the wet season in central Amazonia.

An MLR analysis of optical properties and different species/factors yielded the highest MSE for eBC in all wavelengths followed by the BBOA and AN in the shorter wavelengths (450 and 550 nm), with the addition of AS in the longer wavelengths (637 and 700 nm). Despite having a small mass contribution (6%), eBC dominated the MEE relative contribution (30.8%), followed by the OOA (19.9%). The high MSE of eBC (and a calculated specific SSA of 0.57) is surprising, and more studies concerning the chemical processing of those particles after their emission are needed in order to understand the processes

https://doi.org/10.5194/egusphere-2024-3339
Preprint. Discussion started: 7 November 2024

behind the outstanding efficiency of light scattering of this highly absorbing particle. A special focus should be given to the role of aerosol coating in this process.

While previous studies show a significant contribution of organic particles to light absorption (brown carbon) our study shows that the eBC can also have a significant contribution to light scattering. The quantification of the eBC contribution to light scattering has the potential to improve models and decrease uncertainties in global radiative forcing estimations.

Code availability

Data availability

Data will be made available upon acceptance of the article.

Author Contribution

RS, JFB, and PA conceptualized and designed the methodology. RS performed the field measurements, with the support of JFB. RS, JFB, SC, LVR and JDM applied the methodology. RS wrothe the original draft, and all the authors discussed the results and commented on the paper.

Competing Interests

Some authors are members of the editorial board of ACP.

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Preprint. Discussion started: 7 November 2024

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