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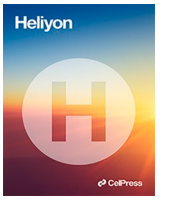

Significant differences in black and brown carbon concentrations at urban and suburban sites

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ABSTRACT

Light-absorbing carbonaceous particles (LAC) may cause and/or exacerbate non-communicable diseases, interfere with the Earth's radiative balance, darken urban buildings and impair vistas. In this study, we explored the temporal behaviour of LAC concentrations measured at wavelengths of 370 nm (brown carbon, BrC) and 880 nm (black carbon, BC) at two sites of a mid-sized city in Brazil. We observed sharp changes in LAC concentrations at the city centre site in response to variations in traffic volume. The highest concentrations were observed when winds originated from both the city core and from the direction of the bus terminal. The suburban site exhibited a notably uniform diurnal pattern and consistently lower LAC concentrations throughout the day. Nevertheless, substantial increases during the evening led to mean BrC and BC concentrations (2.6 and 2.2 μg m^{-3}, respectively) comparable to daytime peaks observed in the city centre (3 μg m^{-3} and 2.5 μg m^{-3}). This phenomenon was attributed to the burning of residential waste and overgrown vegetation in nearby vacant lots. Moreover, the highest concentrations coincided with periods of low wind speeds, usually linked to non-buoyant plumes from point sources. BrC concentrations surpassed BC concentrations, even at the city centre site. Not only was the Ångström absorption exponent ($Å_{370/880}$) larger at the suburban site compared to the city centre (95th percentiles of 1.73 and 1.38, respectively), but it also exhibited a wider span. Overall, the combined LAC and $Å_{370/880}$ data indicated that i) biomass burning is a major source of LAC at the suburban site; ii) at the city centre, bare BC particles may become internally mixed with BrC from biomass or fossil fuel emissions and enhance absorption at lower wavelengths. The occurrence of LAC peaks outside the evening rush hours suggests that other sources but on-road vehicular emissions may contribute to the deterioration of the air quality in the urban core. Tackling air quality across the urban perimeter requires targeting other potential sources but traffic emissions.

1. Introduction

The accelerated pace of climate change observed recently has sparked discussions to tackle the problem from different perspectives, including demand-sided solutions. Such approaches are a critical pathway to reducing greenhouse gas (GHG) emissions, as they include strategies targeting socio-cultural factors (changes in consumption, behaviour, lifestyles), infrastructure use (the design of service provision that enables changes in individual choices and behaviour) and uptake of new technologies by end-users [1]. Besides targeting GHG, climate policies also aim to decrease the concentrations of short-lived climate pollutants, which encompass brown

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carbon (BrC), black carbon (BC), tropospheric ozone and its precursors [2]. BrC and BC particles are emitted from the combustion of carbon containing materials, and exist mostly within the nanometre size range [3]. BrC can also be formed as secondary aerosols linked to humic-like substances (HULIS) through a series of reactions that involve the oxidation of organic compounds (e.g., [4]). Here, BrC and BC will be collectively referred to as light-absorbing carbonaceous particles (LAC) for the sake of brevity. LAC have manifold deleterious effects on the environment: they cause and/or exacerbate non-communicable diseases [5], interfere with the Earth's radiative balance [6], darken urban buildings [7] and impair vistas [8]. Their deleterious effects on human health occur not only because of their small size —which facilitates their penetration in the respiratory system— but also because they act as carriers of co-emitted toxic species that adhere onto their surface. Another important aspect of LAC is that they have been associated with increased risk of cardiopulmonary morbidity and mortality [5,9]. BrC and BC differ in their ability to absorb radiation, with BC strongly absorbing radiation at all visible wavelengths, whilst BrC primarily absorbs radiation at low visible and near ultraviolet wavelengths [10]. This distinction is significant as it has the potential to impact the Earth's radiation balance ([11, 12]).

In urban environments, LAC have traditionally been associated with the combustion of fossil fuels (e.g., vehicular and power plant emissions) and wood burning [13,14]. However, this paradigm has been challenged by a number of studies that showed that the urban sources of LAC are broader and highly dependent on the region, country and the prevailing atmospheric conditions, with complex variabilities between countries, or even between regions in a single country. For example, while vehicular exhaust is a major source of BC in Ljubljana (Slovenia) and Shenzhen (China) [15,16], emissions from wildfire dominate some regions of California during the dry season [17,18] and Scandinavia [19], contributing large amounts of LAC. Krecl et al. [20] reported that the combustion of solid domestic waste in suburban neighbourhoods in a mid-sized Brazilian city led to BC concentrations that exceeded those on busy roads, and Gatari and Boman [21] showed that biomass burning was the main BC source at urban and rural sites in Kenya.

As the average LAC lifespan is relatively short (4–12 days) [22], it is evident that monitoring efforts are required to provide robust understanding of their spatiotemporal distribution, particularly in urban areas where prevailing sources and governing atmospheric conditions are highly variable. Evaluating inter-site variabilities in LAC concentrations necessitates consistent and simultaneous in-situ measurements across multiple receptor sites (for example, as carried out by Zhang et al. [23] at 34 sites in China and Kutzner et al. [24] at 170 sites in Germany). However, access to such vast amounts of data is limited, particularly in the Global South where air quality measurements are either scarce or non-existent. For instance, Brazil, where this study was conducted, lacks a comprehensive, long-term network of LAC measurements. The limited datasets currently available in the country were obtained during short-term campaigns (e.g., [20,25,26]).

Hence, in an effort to fill both scientific and data gaps in this research front, we utilise BrC and BC data collected simultaneously at suburban and urban sites within a mid-sized Brazilian city, wherein we explore their spatiotemporal variabilities. We expand upon

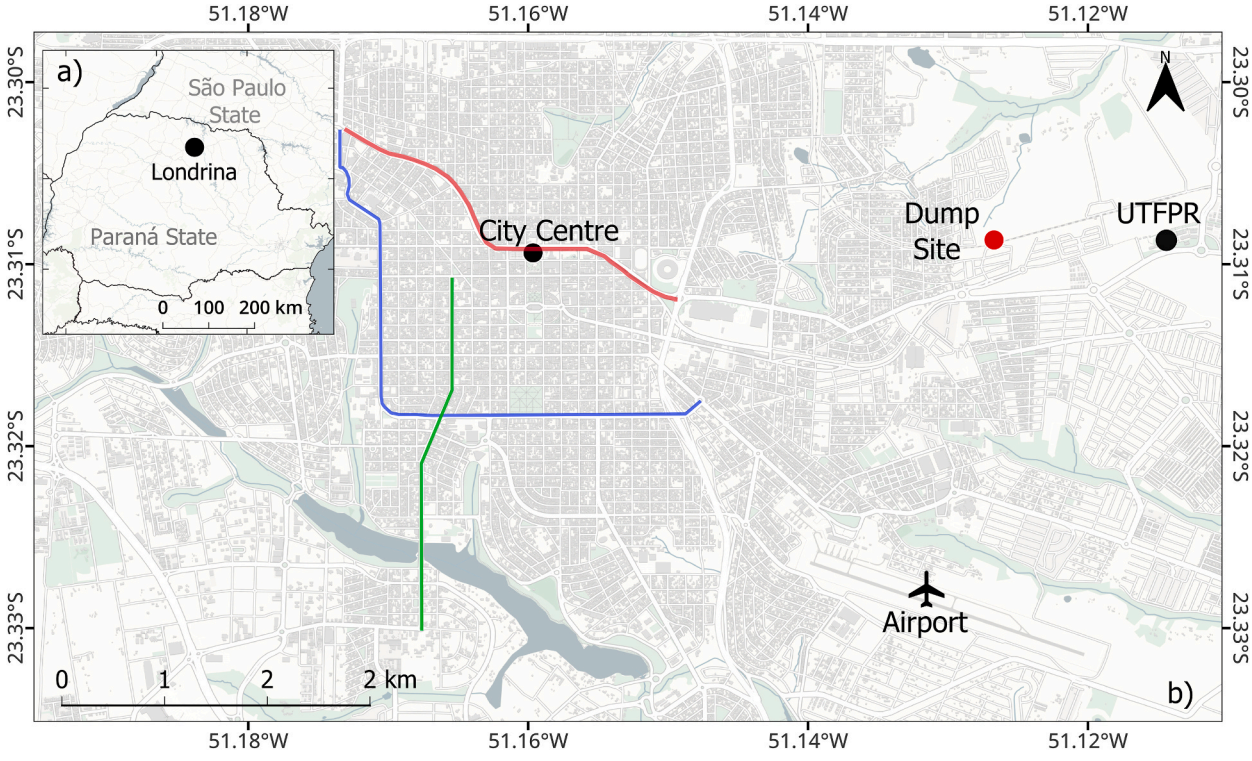

Fig. 1. Maps with the position of Londrina (a) and the study area with the main points of interest (b). The coloured lines in (b) represent the locations of Leste-Oeste Ave. (red), JK Ave. (blue) and Higienópolis Ave. (green). (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

previous works of Krecl et al. [20], Targino et al. [27] and Targino and Krecl [26] and remain consistent with those studies by using many of the same terms, methodology and discussion points.

The primary objective of the study is to identify the potential source areas of LAC and determine the favourable conditions for their transport across the urban perimeter. By analysing inter-site LAC variabilities and pinpointing its sources, the study aims to provide information to aid in the development of local policies designed to alleviate the deterioration of air quality within the city resulting from combustion.

2. Methodology and data analysis

2.1. Study area

We conducted this study at two sites in the city of Londrina (580,870 inhabitants, according to estimates of the Brazilian Census in 2021), located in the state of Paraná, southern Brazil (Fig. 1a). The city has been the subject of numerous studies on urban climate and air pollution, and its main features can be found in Krecl et al. [20], Oukawa et al. [28], Pattinson et al. [29] and Targino et al. [30,31]. Londrina has abundant rainfall between October and February (mean cumulative of 934.5 mm over the period 1976–2019), whilst the rest of the year is drier due to large-scale high pressure systems that block the penetration of rain-carrying fronts, mainly from July to September [27]. This period matches the peak of the wildfire season in central Brazil and neighbouring countries, which significantly impact the air quality of the city.

LAC data were collected from December 3rd, 2015 to January 20th, 2016, embedded in the rainy season. Hence, we assume that the concentrations were not affected by the long-range transport of biomass smoke. However, the burning of domestic waste and grass in vacant lots in some neighbourhoods occur year round [20,32], and affects the local air quality.

2.2. Measurement sites

The LAC mass concentrations were collected in the city centre and on the campus of the Federal University of Technology (UTFPR, located 4.5 km to the east of the city centre) (Fig. 1b). The city centre monitoring site was situated in an area enclosed by roads exhibiting varying traffic densities, and encompassed by the geographical boundaries of Leste-Oeste and JK avenues (Fig. 1b). The road adjacent to the site (Leste-Oeste Ave.) experiences a mean traffic volume of 1080 vehicles h^{-1} during peak hours on weekdays (08:00–09:00 and 17:00–18:00), with 6% constituting heavy-duty vehicles (diesel-fuelled trucks and buses). An additional noteworthy aspect in its location is the proximity to the commuter's terminal, which lies 70 m to the west. This terminal is a transportation hub served by 62 lines, with an average of 2260 diesel-fuelled buses each weekday. The terminal consists of two levels, with the upper level located within an open courtyard enclosed by walls and fences, whilst the lower level is housed inside a roofed building with limited natural ventilation.

The university campus site is classified as suburban due to its location in a neighbourhood with little built-up area and low motorised traffic volume. During the data collection period, the campus was primarily served by a single two-way road in front of the south gate, with a mean traffic volume of 230 vehicles h^{-1} during peak hours on weekdays (08:00–09:00 and 17:00–18:00), of which 5% were classified as heavy-duty vehicles [20].

2.3. LAC measurements

We used two seven-wavelength (λ) aethalometers (model AE-42, Magee Scientific, USA) operated with $PM_{2.5}$ cyclones fitted to the sampling lines, flow rate of 5 L min^{-1}, and sampling rate of 2 min. The sampling lines were installed at 4 m above the ground, 30 m from the road in front the campus, and at 2 m above the ground, 20 m from kerb at the city centre site. The working principle of the aethalometer consists in pumping ambient air through the sampling line into a chamber where particles impact onto a spot of PTFE-coated quartz filter tape. Seven radiation beams (λ = 370, 470, 520, 590, 660, 880 and 950 nm) illuminate the filter, and the instrument continuously monitors the radiation intensity transmitted through a blank portion of the filter and through a spot where particles are deposited. A built-in algorithm uses linear relationships between wavelength-specific absorption cross sections and the radiation attenuation to calculate the LAC mass concentrations [33,34]. LAC measurements at 370 and 880 nm are suitable for environments dominated by biomass and fossil-fuel combustion, respectively. Hence, for the sake of brevity, we focus only on measurements from these two wavelengths and refer to them as BrC (370 nm) and BC (880 nm).

2.4. Calculations

2.4.1. Corrections of LAC data

The linear relationship between the radiation attenuation and LAC concentrations breaks down as the attenuation increases [35]. An artefact called "shadowing effect" occurs as more particles are collected on the filter, in which existing particles obscure those freshly collected. This causes particles to be exposed to different intensity of radiation and to contribute to the attenuation per unit mass in different amounts [34]. Consequently, a non-linear effect arises between the attenuation and LAC concentrations. We used the off-line method proposed by Virkkula et al. [36] to correct the aethalometer data for this effect, in which wavelength- and site-specific compensation parameters are applied to the raw data. Targino et al. [27] used this approach to correct a four-year's aethalometer dataset collected at UTFPR and showed that, if left uncorrected, the LAC mass concentrations may be underestimated by up to 35%.

2.4.2. The Ångström absorption exponent (Å)

The Ångström absorption exponent is a commonly used index for distinguishing the prevalence of aerosols originating from biomass or fossil fuel combustion. Within the scientific community, there is a consensus that fossil fuel combustion generally yields lower Å values compared to biomass burning (lower than 1 *vs.* 1.2–2.5), although there are no universally defined values to establish a sharp cut-off between these sources [37,38]. We calculated Å for the wavelength pair 370 and 880 nm as:

$$\mathring{A}_{370/880} = -\left[\frac{ln\left(\frac{\sigma_{370}}{\sigma_{880}}\right)}{ln\left(\frac{370}{880}\right)}\right], \quad (1)$$

where σ_{370} and σ_{880} are the absorption coefficients at 370 and 880 nm, respectively.

2.4.3. Identification of BrC and BC source regions

The sources of emissions that contribute to both BrC and BC concentrations were identified by analysing wind direction and speed through the use of a conditional bivariate probability function (CBPF) [39]. The CBPF calculates the probability the air pollutant concentrations are larger than specified thresholds for paired wind direction and speed data. The built-in functions of openair R package were used for these calculations [40].

We obtained hourly weather data (air temperature, relative humidity, wind direction and speed, and incoming solar radiation) from Londrina Airport (ICAO: SBLO) meteorological station, located 3.5 km southwest of the UTFPR campus and 4.0 km southeast of the city centre site.

3. Results

3.1. General overview

The weather in Londrina in December and January was typical of summer months, with high air temperatures and abundant rainfall. The mean (± standard deviation) air temperature was 25.2 ± 4.0 °C, relative humidity 77 ± 15% and cumulative rainfall 397.6 mm. The mean values for these variables in December and January (over the period 1976–2019) are 23.9 °C, 74% and 429.7 mm, respectively. The daily solar radiation dose ranged from 2.68 to 28.6 MJ m^{-2}, and it rained on 28 of the 51 days of the experiment, with the largest cumulative 24-hr rainfall of 88.4 mm (coincident with the day of lowest solar radiation dose). The mean windspeed was 3.1 m/s, with these prevailing directions: S (23.7%), SE (17.1%), E (16.3%) and NE (16.1%).

The mean LAC concentrations exhibited distinct patterns at the city centre and UTFPR sites. Specifically, in the city centre, sharp increases in LAC concentrations were observed, with BrC concentrations rising from below 1 before 05:00 h to 3 μg m^{-3} at 07:00 h, and remaining elevated until about 9:00 (Fig. 2a,c). Another increase could be observed starting at 18:00 h. This is a well-studied and established pattern observed at urban sites affected by on-road vehicular emissions, and also reported by Krecl et al. [41] within a street canyon located about 100 m from our city centre site. We attribute the morning peak not only to the increase of vehicular emissions during rush hours, but also to a shallower boundary layer, which inhibits the vertical mixing and facilitates the accumulation of air pollutants close to the ground. At about 9:00 h, the LAC concentrations started to decrease due to the dilution effect of boundary

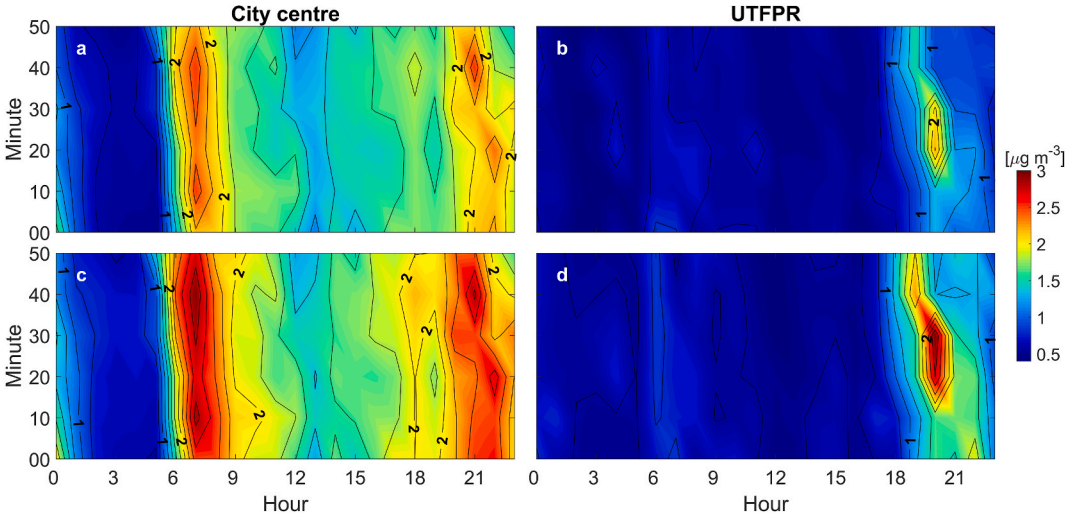

Fig. 2. Time series of 10-min averaged LAC mass concentrations measured in Londrina. a) BC and c) BrC concentrations at the city centre site; b) BC and d) BrC concentrations at UTFPR.

layer growth. As the afternoon progresses and solar radiation diminishes, the transfer of energy from the surface to the atmosphere decreases, resulting in a decline of the boundary layer [42]. However, even though the boundary layer collapses towards the evening, it may still be deeper than in the morning. This phenomenon explains the underlying cause for the lower LAC concentrations at the evening rush hour (BrC = 2.1 μg m^{-3}, BC = 1.8 μg m^{-3} at 19:00 h) compared to the morning (BrC = 3 μg m^{-3}, BC = 2.5 μg m^{-3} at 07:00 h).

An outstanding feature in the LAC concentrations in the city centre is that, compared to BrC, the nocturnal BC hotspot starts later (at about 19:00 h), has a relatively lower peak concentration (2.5 vs. 3 μg m^{-3}) and seems to subsidise at about 23:00 h. The BrC peaks between 21:00 and 23:00 h (i.e., outside the evening rush hour) suggest that the air pollution levels were influenced by other combustion sources other than traffic (for example, waste or biomass burning).

The LAC concentrations at UTFPR remained low throughout the day (Fig. 2b,d). However, at about 18:00 h, a sharp increase created a hotspot that persisted for several hours. Despite being far from major roads, the mean maximum nocturnal BrC and BC concentrations at this site were not negligible (2.6 and 2.2 μg m^{-3}, respectively) and comparable to daytime concentrations in the city centre. Since the UTFPR campus is a suburban site and lacks the large traffic source in immediate proximity, the LAC timeseries also lacks the characteristics commonly associated with emissions from motorised vehicles. Krecl et al. [20] showed that this site is largely impacted by the burning of waste and overgrown vegetation in vacant lots and at a dump site in the late afternoon and evenings. Furthermore, Targino and Krecl [26] showed that BC concentrations peaked at 20:00 h, particularly on days when the air temperature surpassed 30 °C and RH was below 50%. Therefore, we suggest that the hotspots were generated by smoke plumes advected from nearby fires. This hypothesis is further supported by the fact that the BrC signal is more pronounced and covers a more extended time frame than the BC signal. Several observational studies have indicated the significant contribution of biomass burning to BrC mass both at rural and urban sites. For example, Washenfelder et al. [43] reported that the majority of BrC mass at a rural site in Alabama (USA) was associated with this activity. Qin et al. [44] identified BrC as a major absorber of radiation at 370 nm at a suburban site near Guangzhou (China), and Y. Zhang et al. [45] demonstrated that wood burning contributed 18–42% to the total aerosol absorption at 370 nm at nine sites in France. Between 02:00 and 04:00 h, both sites exhibited comparable mean LAC concentrations (ranging from 0.5 to 0.8 μg m^{-3}).

To investigate the temporal variability of LAC concentrations in relation to changes in anthropogenic activities and traffic volume, the datasets were partitioned into two subsets consisting of weekdays and weekends. Because many businesses in Brazil are open on Saturdays, it can be a busy day for shoppers (but not as busy as on weekdays) and, hence, we considered these days as a separate category than Sundays. The category "Sundays" includes not only regular Sundays but also holidays, such as Christmas and New Year's Day.

The application of the Kruskal-Wallis test (at 5% significance level) on the LAC series for weekdays, Saturdays, and Sundays at the UTFPR campus showed no significant statistical differences between weekdays and Saturdays. However, the concentrations on Sundays were significantly different from the other two categories. We observed an increase in LAC concentrations on Sunday evenings, which could be attributed to the tendency of people staying at home and having more time to dispose of their domestic waste. In contrast, when we applied the same test to the city centre concentrations, we found statistically significant differences across all datasets.

BrC and BC concentrations were statistically different at both sites, with BrC consistently exceeding BC, as indicated by all metrics presented in Table 1. This pattern was observed irrespective of the weekday. Specifically, the mean BrC-to-BC ratios were 1.22 and 1.16 at the university and city centre sites, respectively. This finding highlights the relevance of considering BrC as an important contributor to carbonaceous particles in urban environments. Urban studies on LAC pollution often prioritise measurements of BC (most likely due to limitations in instrument capabilities or underexplored datasets); however, our results underscore the need for characterising carbonaceous aerosols at different wavelengths, since BrC can significantly impact air quality in these settings.

Extensive evidence exists that various types of particulate organic material (OM) can absorb radiation in the UV and shorter visible wavelengths (e.g., [46,47]). However, it is important to acknowledge that the aethalometer's signals in this range may not originate

Table 1
Descriptive statistics of LAC concentrations (μg m^{-3}) at the city centre and UTFPR sites.

Site	Statistics	Weekday		Saturday		Sunday	
		BrC	BC	BrC	BC	BrC	BC
City centre	Mean	2.07	1.79	1.78	1.53	1.42	1.17
	Standard Deviation	1.81	1.60	1.52	1.36	1.12	0.89
	Median	1.72	1.50	1.45	1.22	1.09	0.91
	5th percentile	0.37	0.33	0.34	0.30	0.28	0.24
	25th percentile	1.04	0.90	0.95	0.81	0.74	0.64
	75th percentile	2.69	2.30	2.14	1.44	1.78	1.84
	95th percentile	4.84	4.16	3.94	3.39	3.54	2.81
UTFPR	Mean	0.93	0.76	0.82	0.67	0.81	0.68
	Standard Deviation	1.16	1.03	0.69	0.52	1.77	1.69
	Median	0.68	0.54	0.64	0.55	0.50	0.42
	5th percentile	0.16	0.16	0.09	0.13	0.14	0.13
	25th percentile	0.39	0.33	0.34	0.29	0.30	0.26
	75th percentile	1.10	0.87	1.10	0.90	0.98	0.76
	95th percentile	2.29	1.84	1.98	1.46	2.10	1.69

exclusively from BrC particles, but instead they can also be affected by the mixing state of the aerosol. Particularly, when a BC core is coated with OM, the absorption will have contributions from the core and the coating, enhancing the signal as compared to bare BC particles due to the lensing effect [48,49]. This phenomenon may introduce ambiguity in interpretating BC and BrC mass concentrations in environments where OM abounds, such as urban environments. The adsorption of OM onto a BC core can take place in the fumes of internal combustion engines [50,51] or in the atmosphere (e.g., [48]). It is therefore reasonable to expect that a substantial amount of atmospheric BC in urban areas may be internally mixed with OM, thereby enhancing radiation absorption.

3.2. Dependence of LAC concentrations on wind speed and direction

We specifically chose to use median LAC concentrations instead of mean values to represent the relationship with wind patterns, because this metric is more robust and less influenced by extreme values for non-normally distributed data, such as particulate matter [52]. Moreover, in the context of air pollution monitoring, median concentration fields provide a better representation of the average state of the atmosphere, rather than occasional extreme events.

Fig. 3a (BrC) and b (BC) show that the median LAC concentrations at the city centre are dependent on both wind speed and direction. The largest median values occur when winds blow from the west, southwest and northwest at speeds exceeding 6 m/s. This pattern suggests the influence of continuous distant sources that carry pollutants towards the monitoring site, dominating the median LAC concentration field. The sources could be situated in the urban area, since the bulk of the city core lies to the southwest of the receptor site, built on an orthogonal grid plan dominated by motorised traffic. Fig. 3c and d illustrate a substantial reduction in LAC concentrations on Sundays, especially for BC. This finding reinforces that the decrease in traffic volume in the city centre leads to a reduction in carbonaceous aerosol concentrations.

The median BrC and BC concentrations at the UTFPR campus (Fig. 4a and b, respectively) revealed distinct patterns compared to those recorded in the city centre. The concentrations seem to be more evenly spread out across the four quadrants, although a few hotspots stand out during periods of low-speed westerly winds and, to a lesser extent, southerly winds. This behaviour corroborates the presence of intermittent sources in the vicinity, which occur under specific environmental conditions (for example, poor dispersion) and limited time [53]. Easterly winds were nearly completely free of LAC particles.

The CBPF analysis utilising the 75th percentile (P75) revealed that the greatest probabilities of detecting large LAC concentrations at the city centre site are situated within the third quadrant (Fig. 5a). Note that we only show the results for BrC as the patterns of the

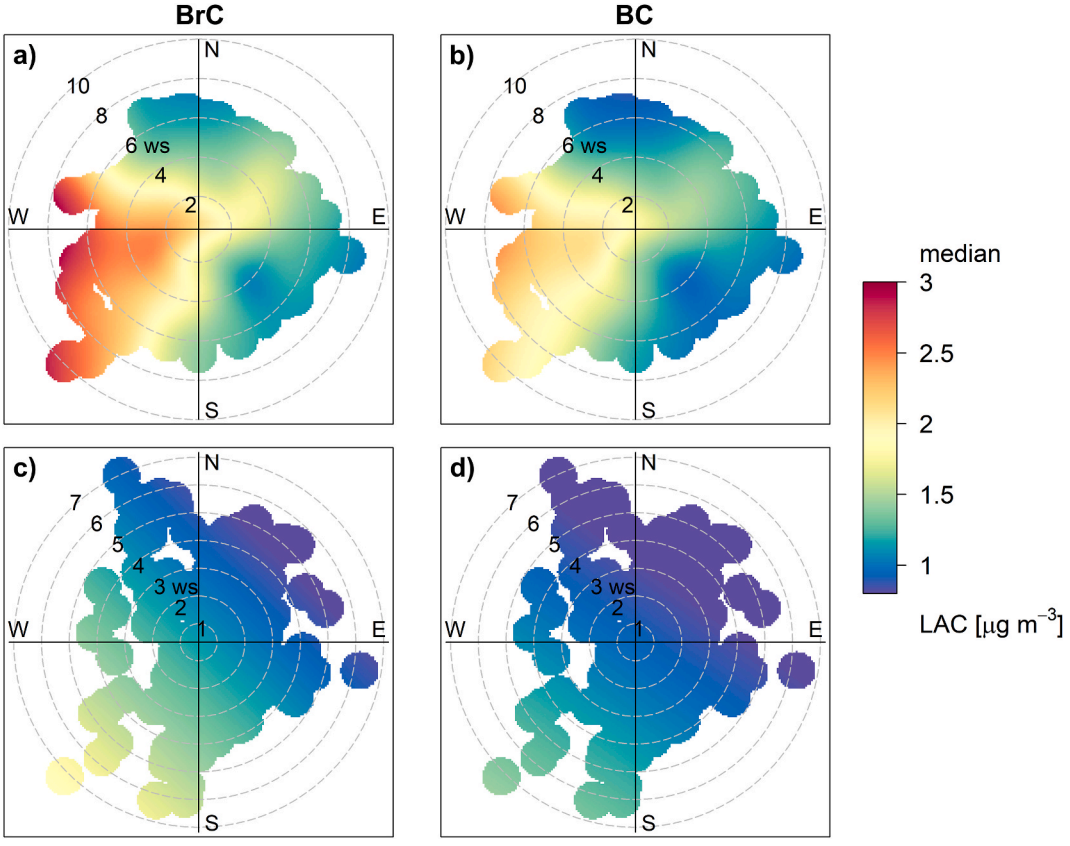

Fig. 3. Polar plots of the median LAC concentrations at the city centre site. Left column: BrC concentrations using all data (a) and Sunday data only (c). Right column: BC concentrations using all data (b) and Sunday data only (d).

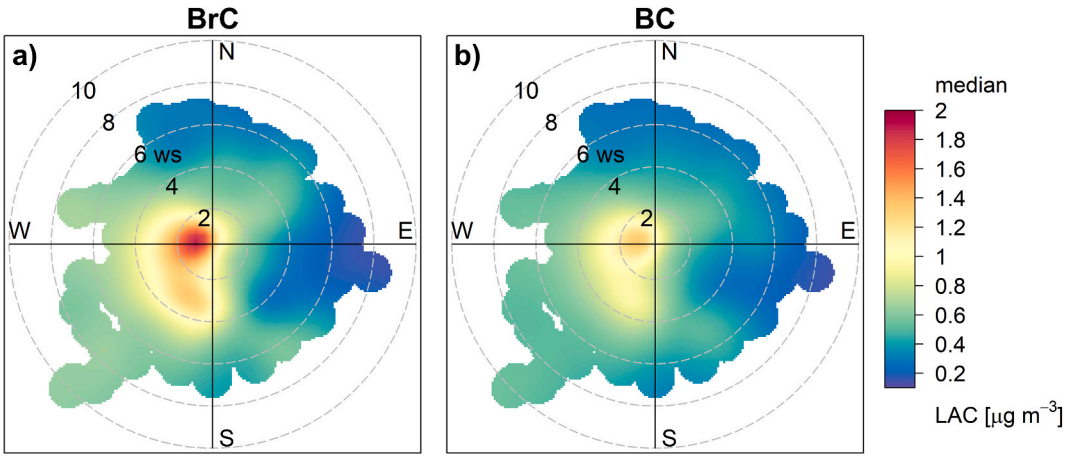

Fig. 4. Polar plots of the median BrC (a) and BC (b) concentrations at UTFPR.

polar plots of BrC and BC are similar. Clearly, the high probability region spans a more restricted area as compared to the median field shown in Fig. 3, but it underscores the significance of southwesterly winds, particularly at speeds below 4 m/s, in enhancing the P75 LAC concentrations (BrC = 2.5 μg m^{-3}; BC = 2.1 μg m^{-3}).

Two busy avenues lie within a radius of approximately 1600 m in the west, southwest and northwest directions: JK Ave. (forming a L-shape configuration) and Higienópolis Ave. (running in the north-south direction). They experience traffic volumes ranging from 980 to 1470 vehicles h^{-1} (between 8:00 and 9:00 a.m.), with an average of 3% consisting of diesel-fuelled heavy-duty vehicles. Under favourable wind directions, these major avenues could potentially contribute to increasing the LAC concentrations at the city centre site.

The low wind speed scenario implies the influence of nearby emission sources. The main likely sources include motorised traffic on

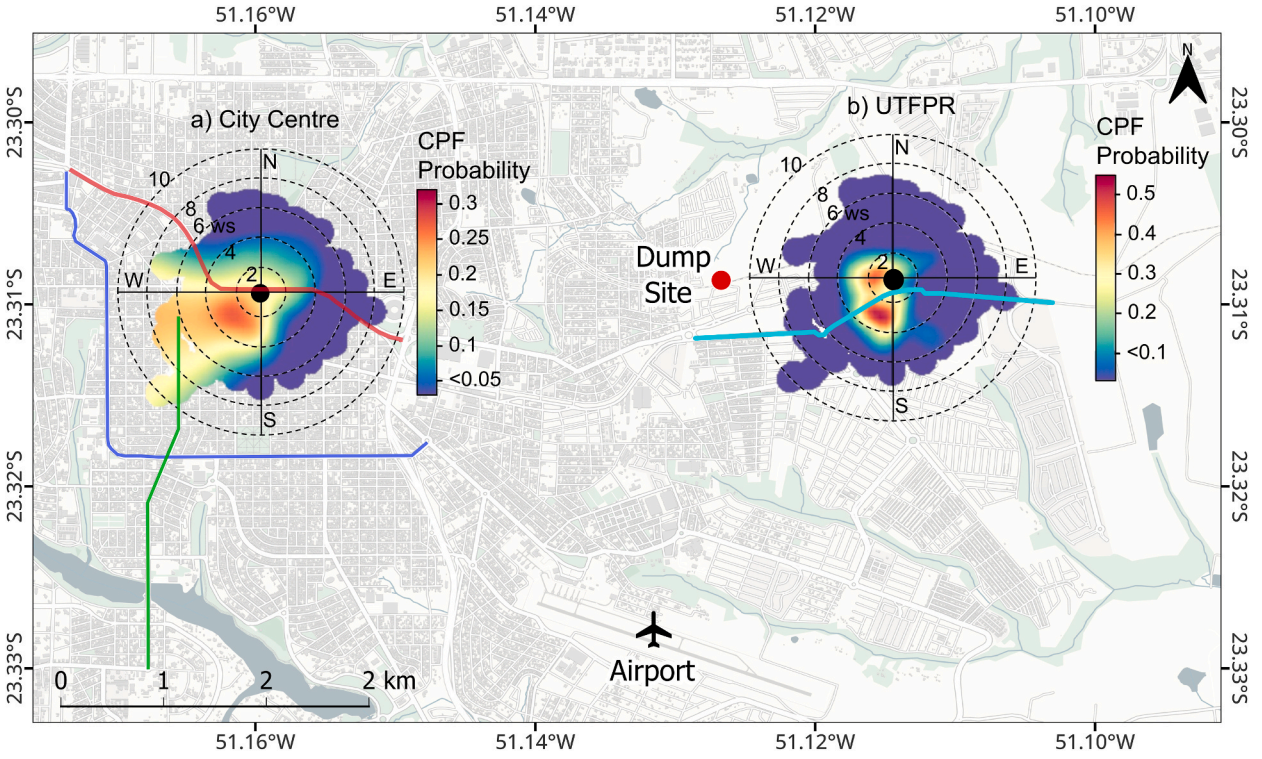

Fig. 5. CBPF plots of BrC concentrations at the city centre (a) and UTFPR (b) sites for concentrations larger than the 75th percentiles (2.1 and 0.85 μg m^{-3}, respectively). The red, blue and green lines in (a) represent Leste-Oeste, JK and Higienópolis Avenues, respectively. The light blue line in (b) represents the road in front the campus' south gate. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

Leste-Oeste Avenue, which runs in an east-west direction with an average traffic flow of 1080 vehicles h^{-1} (the aethalometer's sampling line was placed a few meters away from this avenue), and the city's commuter terminal (located 70 m to the west). The location of the terminal's upper level in an open courtyard allows for natural ventilation, which can help transport pollutants out and towards the receptor site.

On the other hand, the UTFPR campus experienced peak concentrations only under low wind speed conditions, restricted to a limited area extending towards the west and south of the site (Fig. 5b). The region west of the university site has been identified as a potential source of smoke, caused by the burning of overgrown vegetation and waste in vacant lots and at an illegal dump site situated roughly 1200 m away [20]. Additionally, the hotspot to the south of the campus could be linked to motorised traffic in the road that runs parallel to the university's south gate.

3.3. Analysis of the Ångström absorption exponent

It becomes clear from inspecting the Ångström exponent time series (Fig. 6) that the sources of aerosols at the two sites are markedly different. As usually described for sites affected by exhaust from fossil fuels (e.g., [38]), the $Å_{370/880}$ in the city centre were not only lower than those at UTFPR but also exhibited less variability (Fig. 6a,c) over the course of the day. At the latter site, there was considerable variations in $Å_{370/880}$ values, suggesting the presence of different carbonaceous sources.

The lowest $Å_{370/880}$ values at UTPFR occurred between 07:00 and 10:00, which could be attributed to the increase of BC concentrations caused by vehicular traffic entering the campus and circulating on the road in front of the campus. This pattern diluted along the day and transitioned slowly into larger $Å_{370/880}$ values to remain above 1.3 from 18:00 until about 03:00, indicating appreciable contributions from BrC to particle radiation absorption. Whilst we do not have information regarding the use of wood stoves for residential heating or cooking in the area, it is unlikely that the observed LAC concentrations originated from such sources as this is not a common practice in the region. Hence, we attribute these peaks to the enhancement of BrC from open waste and grass burning, as reported by Krecl et al. [20].

Such diurnal transitions in Å values have been observed at other sites with prevalence of both fossil fuel and biomass burning. Dumka et al. [54] found mean nocturnal $Å_{370/880}$ values ranging from 1.20 to 1.23 attributed to emissions from diesel-fuelled trucks in Delhi, but increased to 1.35 in the presence of emissions from residential biomass burning and from open fires in the streets. Similarly, Goel et al. [55] found $Å_{370/880}$ values between 1.13 and 2.42 at an urban site in Delhi, where both fossil fuel and biomass burning sources were present. In a study conducted in Kwadela Township in South Africa, Xulu et al. [56] also reported a substantial diurnal variability in Å values (using the 370/950-nm pair), indicating the fluctuating dominance of emissions from both fossil fuels and biomass burning.

It is worth noting that $Å_{370/880}$ in the city centre increased around 18:00 h, which coincides with the increase observed at UTFPR. We hypothesise that after the evening rush hour, when the traffic volume decreases, the fossil-fuel contribution to LAC subsides, enabling aerosols from biomass burning advected from other areas to dominate the city centre and, as a result, to strengthen the $Å_{370/880}$ signal.

In our study, the $Å_{370/880}$ distribution at UTFPR not only exhibited higher values than in the city centre but also a wider interquartile range (1.05−1.40) (Fig. 6b,d). This implies that there were likely changes in the composition of the aerosols and their optical

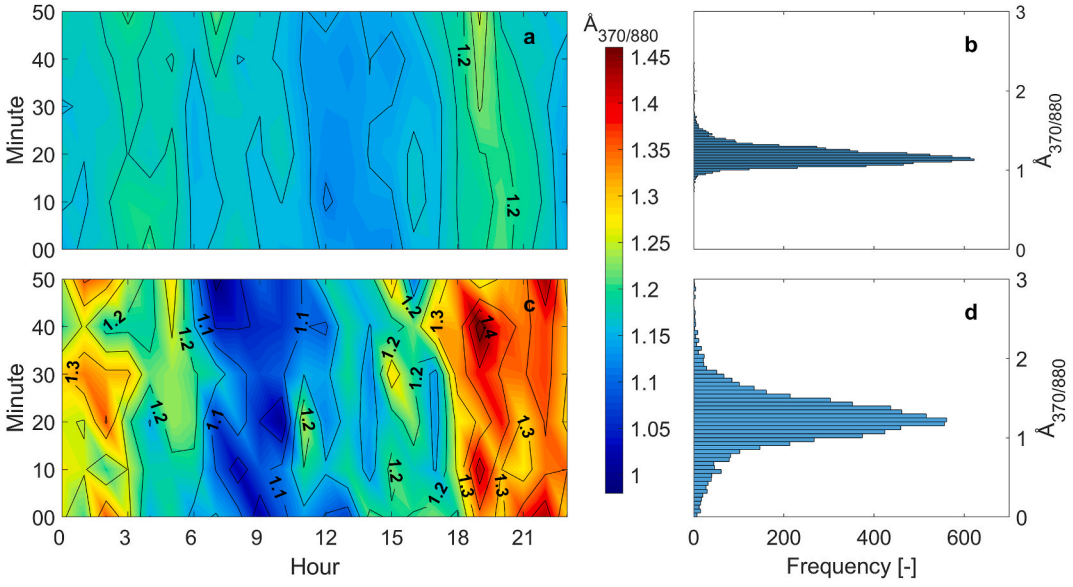

Fig. 6. Time series of Ångström absorption exponent and histograms of distribution for the city centre (a and b) and UTFPR (c and d) sites using 10-min averaged data.

properties, which can occur not only due to changes in the primary sources, but also in the mixing state. For example, particulate OM (including both primary and secondary aerosols) from sources such as biomass and biofuel combustion can become internally mixed with BC [49]. The narrower interquartile range (1.10−1.23) at the city centre site than at UTFPR, with values tightly clustered around a central value, suggests the presence of homogeneous sources from the standpoint of aerosol optical properties. Extreme $Å_{370/880}$ values at the city centre and UTFPR sites in terms of the 95th percentiles were 1.38 and 1.73 using 10-min averaged data.

Overall, relatively large median $Å_{370/880}$ values were observed for all wind directions and speeds, except for strong easterly winds. The city centre site displayed the highest median $Å_{370/880}$ values during high-speed southwesterly winds (Fig. 7a), whilst the campus site presented the highest values over the third quadrant, predominantly during calm winds (Fig. 7b). The use of Å as proxy to separate biomass from fossil fuel contributions or even the commonly assumed value of 1 is debatable (see Gyawali et al. [48], Lack and Cappa [49] and references therein). In addition, more recently Garg et al. [57] found that the combustion efficiency (rather than the fuel used) determines Å, with values approximately equal to 1 for flaming biomass combustion and larger than 1 for older vehicles that operate with poorly optimised engines. Calculations of Å for open waste burning and biomass burning may be equally affected, since they have highly variable combustion efficiencies depending both on environmental variables and conditions of the material (e.g., moisture content). Hence, caution must be exercised when using Å as a proxy for identifying contributions from combustion sources.

4. Limitations of the study

Limited source apportionment: It is important to note that the use of the Ångström absorption exponent as a sole proxy does not offer a conclusive source apportionment without the aid of other combustion markers. Whilst this metric provides valuable insights, it may have limitations in accurately differentiating between fossil fuel and biomass combustion sources. Complementary markers or techniques could enhance the precision of source identification.

Time constraints: The time frame for data collection and analysis was limited in this study. As a result, certain aspects of the research, such as longitudinal data or longer-term effects, were not fully captured.

Sample size: Due to resource constraints, we were only able to include two monitoring sites. A larger sample size could provide more robust and generalisable results in terms of LAC spatiotemporal distribution.

5. Summary and conclusions

This study presented collocated, real-time measurements of light-absorbing carbonaceous particles (LAC): brown carbon (BrC, measured at $\lambda = 370$ nm) and black carbon (BC, at $\lambda = 880$ nm). We explored the temporal behaviour of LAC concentrations at two sites within the urban area of Londrina, a mid-sized city in southern Brazil. Our results clearly showed pronounced spatiotemporal variability in both BrC and BC concentrations related to on-road motorised traffic and biomass burning emissions. At the city centre site, the sharp transitions in LAC concentrations occurred in response to the variability in traffic volume. The BrC concentrations were found to be significantly higher than the BC concentrations. The occurrence of nocturnal LAC peaks outside the traffic rush hours and the BrC:BC ratio larger than 1 in the city centre suggest that on-road vehicular emissions may not be the main culprit of poor air quality in the urban core at night (for example, smoke from biomass burning may contribute substantial amounts of LAC). This also seems to challenge the paradigm that BC dominates as the primary carbonaceous component in traffic-dominated environments. This result underscores the need for greater attention to be given to BrC, which can impact air quality in these settings.

LAC concentrations at the suburban site peaked in the evening and remained elevated for a few hours. Although the use of the Ångström absorption exponent does offer a conclusive source apportionment without the aid of other markers, we suggest that the prevalence of relatively large $Å_{370/880}$ values in the evening is indicative of biomass burning. Moreover, the conditional bivariate probability function analysis indicated that the large BrC concentrations originated from local sources. Based on our postulations and previous observations in the area, we propose that the burning of residential waste and grass were the main fuels contributing to this phenomenon. Whilst the suburban site consistently exhibited low LAC concentrations during the daytime, the persistent nocturnal peaks observed raise concerns. These peaks are worrisome not only due to the cumulative health risks associated with exposure to high concentrations, but also because the fire foci occur in socially vulnerable communities already suffering from environmental injustices.

Our study underscores the importance of incorporating multiple wavelength LAC measurements in future research aimed at characterising carbonaceous aerosols and their potential combustion sources. However, it should be noted that the availability of LAC measurement capabilities is not always widespread, especially in the Global South. Therefore, a collaborative effort within the aerosol community is crucial to expand the coverage of LAC measurements in areas where they are scarce or non-existent. Furthermore, a reorientation of traditional approaches to address urban air pollution is required to effectively tackle the complexities of urban air quality and to target other potential sources but traffic emissions.

Author contribution statement

Admir Créso Targino: Conceived and designed the experiments; Performed the experiments; Analyzed and interpreted the data; Wrote the paper.

Fabian Leonardo Moreno: Analyzed and interpreted the data; Contributed reagents, materials, analysis tools or data.

Patricia Krecl: Conceived and designed the experiments; Analyzed and interpreted the data; Wrote the paper.

João Vitor Cardoso: Analyzed and interpreted the data; Contributed reagents, materials, analysis tools or data.

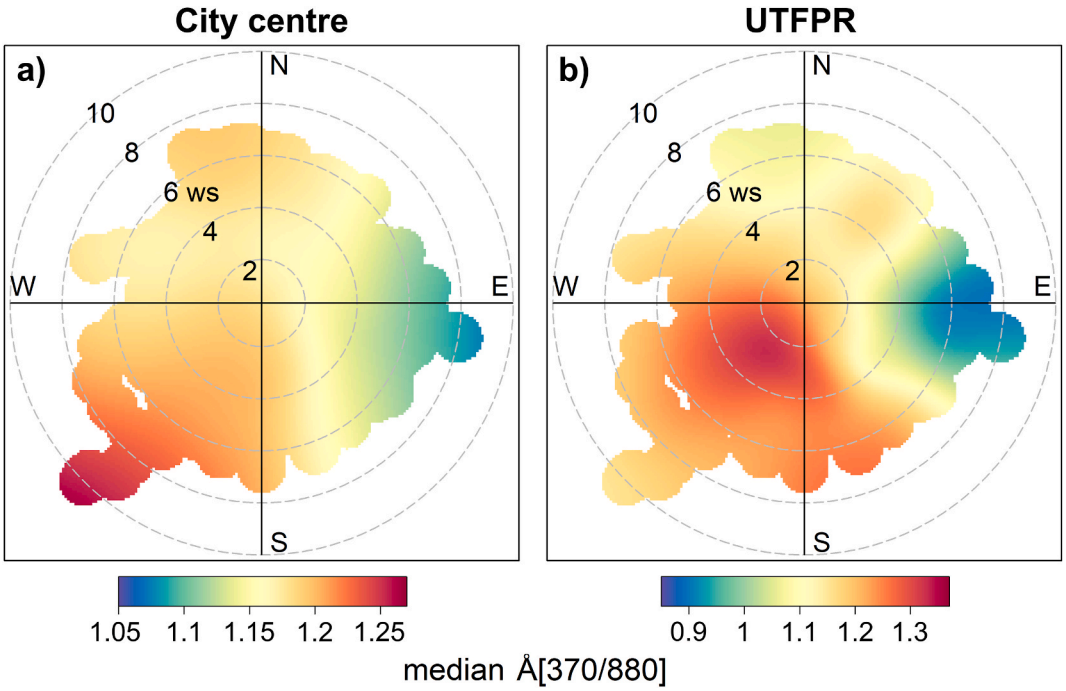

Fig. 7. Polar plots of median $Å_{370/880}$ at the city centre (a) and UTFPR (b) sites. Note that different scales were intentionally utilised to accentuate the distinct features.

Data availability statement

The authors do not have permission to share data.

Additional information

No additional information is available for this paper.

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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