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Three-year Measurements of Black Carbon Concentrations in Sofia, Bulgaria

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Abstract: A three-year continuous monitoring campaign of black carbon (BC) concentrations was conducted at an urban background site in Sofia, Bulgaria, from February 2022 to December 2024. BC concentrations were measured with an aethalometer (Model AE-33, Magee Scientific, USA). The main objective was to assess the temporal variability of BC concentrations, quantify the contributions from fossil fuel and biomass burning sources, and explore the relationships between BC levels and meteorological parameters such as wind speed and temperature. Hourly BC concentrations ranged from 0.07 to 24.94 μg/m³, with a mean ± s.d of 2.96 ± 2.49 μg/m³. Daily average BC ranged from 0.5 to 12.75 μg/m³, with a mean ± s.d 2.96 ± 1.86 μg/m³. A distinct bimodal distribution of BC concentrations was observed across all seasons during rush hour traffic, with a morning peak between 10:00-11:00 and an evening peak around 20:00-21:00. Fossil fuel combustion, which main source are diesel vehicles was the dominant source of BC at Sofia site across all seasons, contributing on average ~ 91% of total BC (ranging from 92% in summer to 84% in winter). The percentage contribution of biomass burning to BC (BB%) - from sources such as residential heating, agricultural burning, and wildfires - was most pronounced in winter. BB% followed a clear seasonal pattern, with mean values 16% in winter, 11% in spring, 10% in autumn, and 8% in summer. BC concentrations showed an inverse relationship with both wind speed and temperature. Higher BC levels were associated with calm and colder conditions, consistent with typical urban pollution dynamics, where stagnant air favors pollutant accumulation, and higher wind speeds and temperatures promote dispersion and dilution.

Keywords: Black carbon, source apportionment, AE-33, air quality, urban atmosphere

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1. INTRODUCTION

Black carbon (BC) is a major component of fine particulate matter ($PM_{2.5}$) produced through incomplete combustion of fossil fuels, biofuels, and biomass (Bond et al., 2013). It is associated with serious impacts on climate change and human health.

Due to its small size BC can penetrate deep into the respiratory system, carrying with it toxic compounds into the bloodstream. Numerous epidemiological studies have found strong associations between both short- and long-term exposure to BC and increased risks of cardiovascular and respiratory diseases (WHO, 2012; Zhu et al. 2023). In urban populations, where combustion sources are concentrated, BC exposure has been linked to elevated hospital admissions and premature mortality (WHO, 2012).

From a climate perspective, BC warms the atmosphere because it is very efficient at absorbing solar radiation. This heat absorption contributes to atmospheric warming, which makes BC one of the most important contributors to atmospheric warming after carbon dioxide, even though it has a much shorter lifetime compared with CO_2 (Ramanathan&Carmichael, 2008). Both the immediate health risk and short-term climate forcing makes BC a priority target for mitigation strategies aimed at delivering rapid co-benefits for air quality and climate (Shindell et al., 2012).

The new European Ambient Air Quality Directive (NAQD), adopted in October 2024, requires monitoring of new pollutants such as ultrafine particles (UFP), particle number size distributions (PNSD), black carbon (BC), elemental carbon (EC), ammonia (NH_3), multiple volatile organic compounds (VOCs), and oxidative potential (OP) of PM in order to support scientific understanding of their effects on health and the environment (Savadkoohi et al., 2024).

Monitoring of air pollutants, as well as the determination of their sources and how they change over time are essential for effective air quality management and mitigation efforts.

Major sources of BC include vehicles (particularly diesel-driven road vehicles), non-road mobile machinery (e.g., forest machines), ships, residential heating (e.g., small coal or wood-burning stoves), and open biomass burning (e.g., forest fires or the burning of agricultural waste) (EEA, 2013). These sources can vary significantly between regions, making reliable measurement methods essential for accurate assessment.

BC concentration cannot be measured directly by weighing collected aerosol particles; instead, it is determined indirectly, most commonly using optical methods. (Bond et al, 2013). A widely used are filter absorption photometers (FAPs), which measure the attenuation (σ_{ATN}) or transmission of light through a particle-loaded filter tape and convert them to the absorption of light (Savadkoohi et al, 2023). One of the most commonly used FAPs instrument is the Aethalometer (AE), which estimates BC concentrations from the amount of light absorbed by the particles collected on filter tape samples. The BC measurements can be further analyzed using the Aethalometer model (Sandradewi et al., 2008), which distinguishes between two source categories - fossil fuel combustion (BCff) and biomass burning (BCbb) - based on their different wavelength dependencies of light absorption by aerosols.

BCff originates from the incomplete combustion of fossil fuels such as diesel, coal, gasoline, and natural gas and its main sources are vehicles (especially diesel engines), industrial processes, and power plants. BCbb results from the burning of organic material, such as wood, agricultural waste, and forest fires, with main sources being residential heating, agricultural burning and wildfires. The percentage contribution of biomass burning to BC (BB%) is the proportion of BC originating from biomass burning sources.

Numerous studies across Europe consistently show that fossil fuel combustion is the dominant source of ambient BC in urban areas, whereas biomass burning contributes more significantly to BC levels in rural and suburban regions, particularly during the cold months (Sandradewi et al., 2008; Mousavi et al., 2019; Helin et al, 2018; Briggs&Long 2016; Titos et al, 2017; Diapouli et al, 2017). In eastern and southern Europe BC concentrations are higher compared to northern Europe, which reflects patterns seen with other air pollutants. There is also a lack of monitoring of BC at traffic sites in these regions (Savadkoohi et al., 2024).

Measuring campaigns on BC concentrations in Bulgaria are limited in number and are not conducted regularly. Only a few studies on monitoring BC concentrations in $PM_{2.5}$ emerged in Bulgaria in the past 5 years, in which daily samples of $PM_{2.5}$ on filters are evaluated for BC concentrations and source apportionment obtained with the Multi-wavelength Absorption Black instrument (MABI). (Hristova et al, 2022; Hristova&Veleva, 2020; Hristova et al, 2021) The MABI is a research instrument, it is very flexible but in order to calculate BC values it requires user support and custom processing, making it not suitable for long term, unattended monitoring, especially for high resolution data (Manohar et al, 2021; Hristova et al, 2022). The Aethalometer on the other hand is more appropriate for unattended long term BC monitoring - with built-in corrections and source apportionment models.

Sofia, as the capital and largest urban center in Bulgaria, with 1.2 million inhabitants (NSI), is a particularly relevant location for such monitoring. It is located in a semi-closed valley, providing favorable condition to strong temperature inversions, mainly in autumn and winter (Hristova et al, 2021). It experiences high levels of air pollution, particularly in cold season due to a combination of meteorological conditions, the topography of the city, urban traffic congestion and increased use of solid fuels for heating in cold season.

To address the lack of long-term data in this area, we conducted a three-year continuous monitoring campaign of BC concentrations at an urban background site in Sofia. An aethalometer (Model AE-33, Magee Scientific, USA) was used to measure BC concentrations from February 2022 to December 2024 (Magee Scientific, 2016). The main objective was to assess the temporal variability of BC concentrations, quantify the contributions from fossil fuel and biomass burning sources, and explore the relationships between BC levels and meteorological parameters such as wind speed and temperature.

2. METHODS AND DATA

From February 2022 to December 2024 an aethalometer (Model AE-33, Magee Scientific, USA) was used to measure BC concentrations at an urban background site in Sofia. An air inlet (2.5 µm cut at 2 LPM) was installed at 5.5 m a.g.l. The AE-33 continuously collects aerosol particles by passing the aerosol-laden air stream through the filter tape (M8060). BC concentrations are estimated from the amount of light absorbed by the particles collected on the filter tape samples. The optical attenuation is measured on two spots with different sample flows and on a reference spot without flow with high time resolution of 1s or 1 min (Drinovec et al, 2015). Measurments are performed at seven optical wavelengths (λ = 370, 470, 520, 590, 660, 880 and 950 nm), ranging from near infrared to near ultraviolet (Magee Scientific, 2016). The change in optical attenuation at 880 nm is used to calculate the BC mass concentration, because at this wavelength, other aerosol particles, mostly organic aerosols, absorb only a negligible amount of light, and the absorption can be attributed almost entirely to BC. (Drinovec, 2015)

The AE33 uses the dual-spot method to automatically correct in real time for the accumulation of particles on the filter tape (filter loading effect). It also applies a multiple-scattering correction using the parameter C0, which for the M8060 filter tape is predefined as 1.39 based on manufacturer calibration (Savadkoohi et al, 2024). No additional post-processing or offline corrections were applied to the BC data in this study.

The source apportionment of BC into fossil fuel (BCff) and biomass burning (BCbb) fractions is performed using the Sandradewi et al. (2008) Aethalometer model, which assumes that the total optical absorption coefficient can be expressed as the sum of these two source contributions. The model applies optical parameters that describe the spectral dependence of aerosol light absorption, known as absorption Ångström exponents (AAEs) (Savadkoohi et al, 2024). By default, the AE33 instrument uses αff = 1 for fossil fuel and αbb = 2 for biomass burning (Magee Scientific, 2016; Sandradewi et al., 2008). These values are not directly measured but are assumptions in the model that can be adjusted based on local conditions; in this study, the default values were used.

Using these assumptions, the BC mass concentration and its source contributions were derived using the following equations.

Black carbon concentration is based on the formula:

$$BC = \frac{b_{abs}}{\mathrm{MAC}},$$

where b_{abs} is absorption coefficient, MAC is mass absorption cross-section, measured in $m^2\ g^{-1}$. The MAC represents the absorption efficiency of BC particles, it is highly variable and is influenced by different factors such as the site location (e.g., urban, rural, high-altitude), BC particle size, internal vs external mixing, combustion sources and instrument type (Savadkoohi et al, 2024). We used the nominal MAC values from instrument manufacturers of 7.77 m² g⁻¹, because we haven't yet estimated the site-specific MAC for Sofia site.

The percentage contribution of biomass burning to BC (BB%) - the proportion of BC originating from sources such as residential heating, agricultural burning, and wildfires is obtained from the AE33 by the following formula:

$$BB\ (\%) = \frac{b_{abs}(950\ nm)_{bb}}{b_{abs}(950\ nm)}\ ,$$

where $b_{abs}(\lambda)$ is absorption coefficient, λ=950 nm is wavelength, $b_{abs}(\lambda)_{bb}$ a biomass burning fraction of absorption coefficient.

Biomass burning (BC_{bb}) and fossil fuel (BC_{ff}) fractions are then calculated as:

$$BC_{bb} = BB * BC,$$
$$BC_{ff} = (1 - BB) * BC$$

Data evaluation

The BC measurements were obtained by the Aethalometer at a time resolution of 1 min. We computed hourly average values by aggregating the one-minute data into 60-minute means for each hour. BC concentrations were originally recorded in nanograms per cubic meter (ng/m³) and were converted to micrograms per cubic meter (µg/m³) for consistency with common reporting standards.

Due to technical issues, the Aethalometer was out of operation during certain periods of the three-year monitoring campaign. These interruptions occurred in different months and were not evenly distributed over time. Table 1 shows the number of days with available BC measurements for each year and season. Seasons are defined as: spring: March-May; summer: June-August; autumn: September-November; winter: December-February. In spring, data were available for 259 days, while in winter 106 days were recorded. Gaps in the dataset are due to intermittent Aethalometer downtime, unevenly distributed across months. This uneven distribution should be kept in mind when comparing seasons, since winter is underrepresented.

Table 1. Number of days with available BC measurements per season and year

	Spring	Summer	Autumn	Winter
2022	92	5	25	14
2023	79	49	59	45
2024	88	90	91	47
Total	259	144	175	106

The meteorological parameters (wind speed and temperature) were measured simultaneously with BC monitoring at NIMH station in close proximity (200m) from the location of the aethalometer.

The collected data on BC concentrations, along with the meteorological parameters were analyzed using the statistical software R 4.5.1 (R Core Team, 2025) and the dplyr (Wickham, 2023), lubridate (Grolemund&Wickham, 2011) and ggplot2 (Wickham, 2016) packages.

3. RESULTS

To assess the temporal variability of the hourly BC concentrations and BB% during the period Feb 2022 – Dec 2024, hourly, daily (day of week) and monthly patterns were examined using box plots (Fig. 1). The highest BC concentrations were observed in colder months (November, December and January), on weekdays and around 10:00-11:00, while the concentrations were lowest in summer months, on weekends and during night hours (top panel, Fig. 1). The hourly BC concentrations ranged from 0.07 to 24.94 µg/m³, with a mean ± s.d of 2.96 ± 2.49 µg/m³. The daily average BC ranged from 0.5 to 12.75 µg/m³ with a mean ± s.d 2.96 ± 1.86 µg/m³.

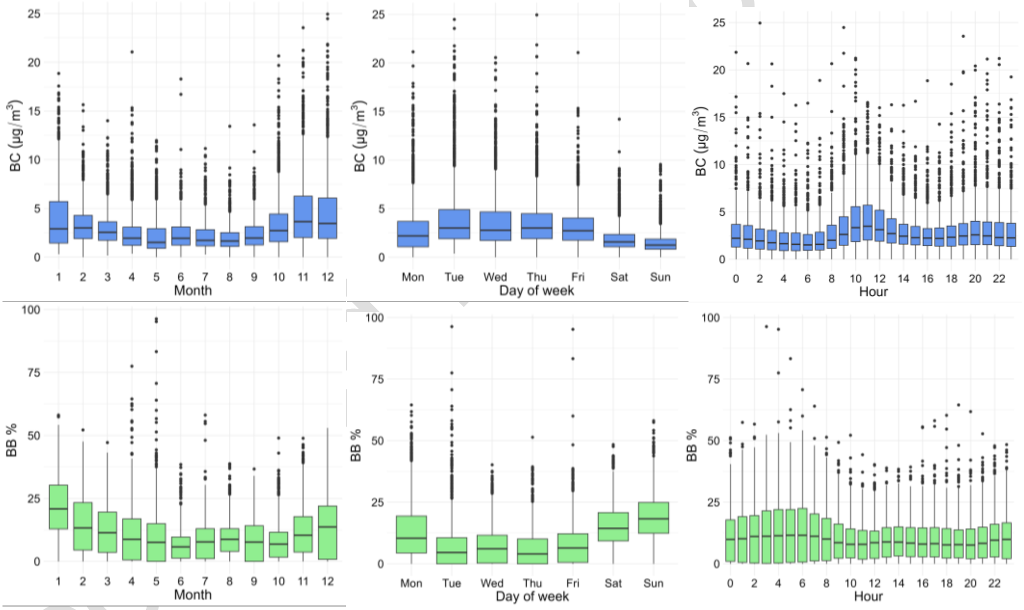

Fig. 1. Temporal variability of monthly, daily (day-of-week), and hourly patterns of BC concentrations (top panel) and BB% (bottom panel), based on hourly data.

BB% was highest during winter, on the weekends and at nighttime hours, while in summer months it showed the lowest impact (bottom panel, Figure 1). The presence of significant outliers in spring likely suggests that occasional biomass burning events (e.g., wildfires, agricultural burning) contribute to short-term pollution spikes.

Table 2 summarizes the descriptive statistics of BC, BCff, BCbb concentrations and BB% over the entire monitoring period and by season. The average BC concentration was 2.96 μg/m^3, with a clear seasonal pattern: lowest in summer (2.13 μg/m^3) and highest in winter (4.07 μg/m^3). Fossil fuel combustion (BCff) was the dominant source year-round, contributing about 91% of total BC, with mean concentrations ranging from 2.0 μg/m^3 in summer to 3.48 μg/m^3 in winter. Biomass burning (BCbb) contributed around 9% of total BC when expressed as a mass fraction, while the mean of hourly BB% values was 10.7%. Both indicators showed a pronounced seasonal increase during winter (to about 16%), consistent with enhanced residential wood burning during the heating season. It should be noted that 142 hourly BC values (< 1% of the total 16175 hours) and 2573 hourly BB% values (~ 16%) were recorded as zero or below the instrument's detection limit. Therefore, the minimum values reported in Table 2 correspond to the second-lowest observed values above zero.

Table 2. Descriptive statistics (mean, minimum [Min], and maximum [Max] concentrations) of BC, BCff (fossil fuel BC), BCbb (biomass burning BC), and BB % based on hourly data measured from Feb 2022 to Dec 2024

Variable	Entire period	Spring	Summer	Autumn	Winter
N (days)	684	259	144	175	106
N (hours)	16175	6200	3346	4138	2491
	Mean [Min-Max]				
BC (*μg/m^3*)	2.96	2.49	2.13	3.66	4.07
	[0.07-24.94]	[0.07-21.06]	[0.27-18.28]	[0.1-23.55]	[0.24-24.94]
BCff (*μg/m^3*))	2.7	2.29	2	3.4	3.48
	[0.03-24.47]	[0.03-21.06]	[0.176-18.28]	[0.08-22.94]	[0.15-24.47]
BCbb (*μg/m^3*))	0.26	0.2	0.13	0.26	0.59
	[0.0007-6.86]	[0.0007-3.31]	[0.0009-1.7]	[0.001-3.02]	[0.002-6.86]
BB (%)	10.73 %	10.66 %	8.32 %	9.65 %	15.95 %
	[0.1-96.3]	[0.1-96.3]	[0.1-58.1]	[0.1-49]	[0.1-58.1]

The hourly average BC changes throughout the day for each season are shown in Fig. 2 (left). A distinct bimodal distribution of BC concentrations was observed across all seasons during rush hour traffic, with a morning peak between 10:00-11:00 and an evening peak around 20:00-21:00. The lowest BC concentrations consistently occurred in the early morning hours, between 05:00-06:00. The shape and timing of the BC (red) and BCff (blue) curves almost fully overlap, especially in spring, summer and autumn, suggesting that BC is primarily from fossil fuel sources. In winter there is a bigger gap between those two curves, indicating more significant contribution from biomass burning sources, which can also be seen from the BCbb curve (green) which increases slightly in winter. The hourly average BB% throughout the day for each season are displayed on the right side in Fig 2. BB% exhibited a clear seasonal pattern with highest hourly average percentages during winter (20%), followed by spring and autumn (13%)

and lowest during summer (9%). The highest BB% occurs at nighttime hours between 02:00-06:00 during all seasons.

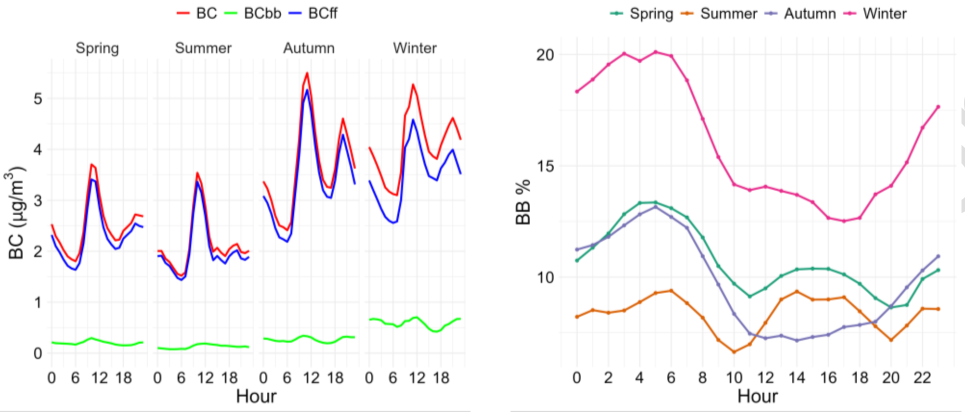

Fig 2. Seasonal variation of BC (black carbon), BCbb (biomass burning BC), BCff (fossil fuels BC) (left), and BB% (biomass burning proportion) (right)

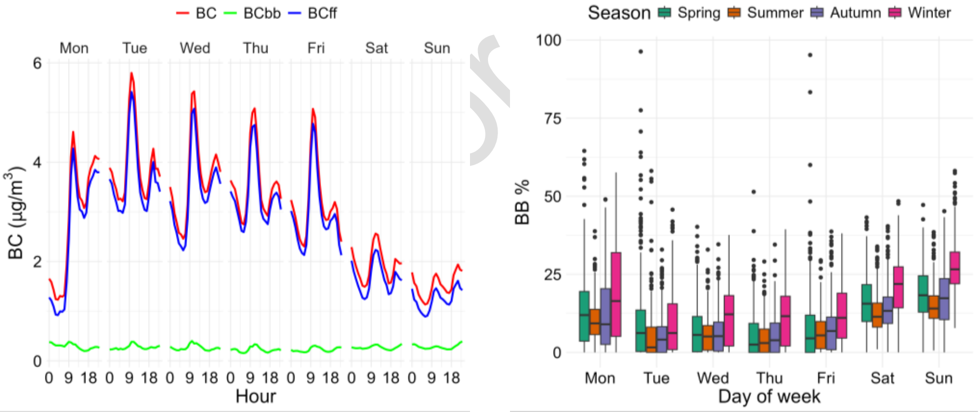

Fig. 3. Diurnal (day of week) variation of BC (black carbon), BCbb (biomass burning BC), BCff (fossil fuels BC) (left), and box plot of the seasonal variation of BB% by weekday (right)

The diurnal variation of BC, BCbb and BCff is shown in Figure 3, left. The plot illustrates a distinct diurnal pattern in BC concentrations, with peak levels during typical rush hour traffic on weekdays, driven largely by fossil fuel combustion (BCff). Saturday and Sunday show significantly lower BC and BCff concentrations compared to weekdays. The morning and evening peaks are still present on weekends but dampened, reflecting reduced traffic and potentially changes in human activity.

BCbb (green line) remain relatively low and show little variation across hours and days. A slight increase is observed during morning or evening hours, but its contribution is clearly minor compared to BCff. This further confirms that biomass burning is a secondary source at this urban site, and fossil fuel emissions dominate.

Box plot of the seasonal variation of BB% by weekday is displayed on the right side in Fig 3. BB% vary strongly by season and day of the week. Winter shows the highest BB% values, particularly on weekends, where medians are around 25%, compared to 12% on weekdays. Spring and autumn show moderate contributions, while in summer BB% remains consistently low. Across all seasons, Saturday and Sunday have higher BB% than on weekdays.

In summary, biomass burning is most intense in winter, especially during weekends, likely due to residential heating (especially in houses using wood or coal) and less work-related traffic emissions, leading to higher BB% by proportion.

In Figure 3 (right), Mondays across all seasons show slightly higher BB% values compared to other weekdays, likely reflecting a carryover from weekend biomass burning activities. This pattern is also visible in Figure 3 (left), where greater variability is observed from Sunday evening into Monday morning. The effect is further illustrated in Fig. 4, which compares daytime (08:00-20:00) and nighttime (20:00-08:00) BC and BB% values.

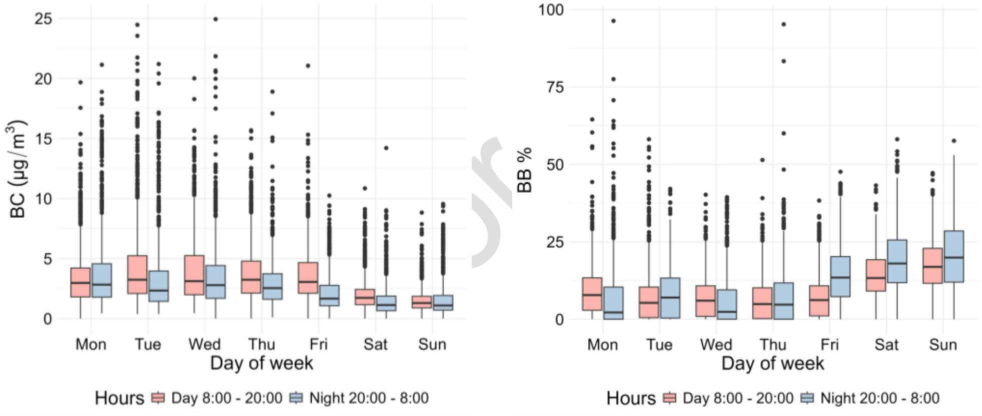

Fig. 4. Boxplots comparing daytime hours (8:00-20:00) in red vs nighttime hours (20:00-8:00) in blue for BC (left) and BB% (right)

The boxplot in Figure 4 (left) shows that BC concentrations are generally higher during daytime hours, consistent with traffic-related emissions. In contrast, Figure 4 (right) demonstrates that BB% is markedly elevated at night during weekends (Friday-Sunday), reflecting increased residential wood burning, whereas on weekdays daytime and nighttime values remain relatively similar.

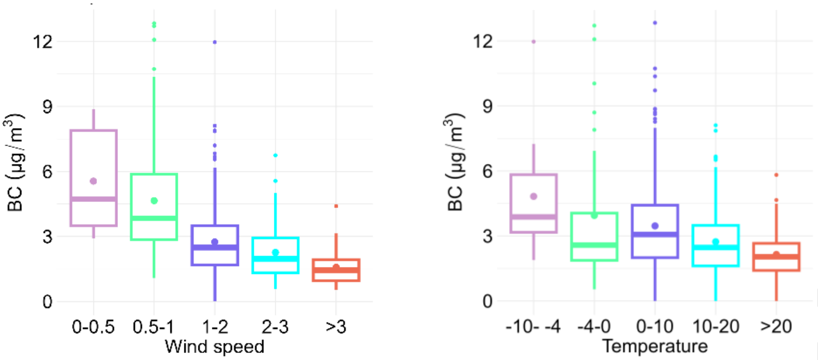

Fig. 5. Box plots of daily average BC concentrations grouped by wind speed (left) and temperature in °C (right) and divided into 5 ranges

The influence of wind speed on BC concentrations was evaluated through box plot visualization (Figure 5, left). The plot shows an inverse relationship between daily average BC concentrations and wind speed. BC concentrations differed significantly among wind speed categories (Kruskal-Wallis, $\chi^2 = 121.6$, df = 4, $p < 0.001$). Post-hoc Wilcoxon tests with Bonferroni correction confirmed significant differences between all groups, except between 0-0.5 and 0.5-1 m/s, which did not differ. The BC concentrations were higher under low wind conditions and lower during windier periods. This aligns with typical urban air pollution dynamics, where stagnant air conditions trap pollutants, while higher wind speeds help to disperse them. Similar results were observed for BC concentrations in relation to temperature (Fig. 5, right). BC concentrations varied significantly with temperature (Kruskal-Wallis, $\chi^2 = 58.7$, $p < 0.001$). Post-hoc tests showed that concentrations were consistently higher at $T < 10$ °C compared to $T > 20$ °C ($p < 0.001$), while no significant differences were observed among the colder categories below 10 °C. Lower air temperatures require more building heating. In addition, when $T < 10$ °C, traffic emissions rise due to the cold start of engines. BC concentrations remained high and relatively stable across the cold temperature categories but decreased progressively with increasing temperature, reaching the lowest levels during warm periods (>20 °C).

Previous measurements at the same urban background site, conducted between June 2020 and July 2021 on daily $PM_{2.5}$ samples using a Multi-wavelength Absorption Black instrument (MABI), showed mean monthly BC concentrations ranging from 1.35 to 4.42 µg/m^3 (Hristova et al., 2022). The lowest mean monthly $PM_{2.5}$ concentrations were observed in May 2021, while the highest occurred in November 2020.

Overall, the BC levels measured in Sofia are higher than those typically reported for Western and Northern Europe and for rural areas (Liu et al., 2023; Zanatta et al., 2016; Savadkoohi et al., 2023; Cesari et al., 2025). In contrast, the measured BC concentrations in Sofia, both as daily means and peak values, are lower than those reported for several Asian cities (Goel et al., 2020; Dutkiewicz et al., 2009). These differences may be explained by the size of the urban area, specific meteorological conditions, and the age and type of vehicles in use.

4. CONCLUSION

A continuous measurement campaign of black carbon (BC) concentrations was conducted at an urban background site in Sofia, Bulgaria, from February 2022 to December 2024, using an Aethalometer (Model AE-33, Magee Scientific, USA). The analysis of BC temporal variability shows that hourly BC concentrations ranged from 0.07 to 24.94 µg/m^3, with a mean ± s.d. of 2.96 ± 2.49 µg/m^3, while daily average BC ranged from 0.5 to 12.75 µg/m^3, with a mean ± s.d. of 2.96 ± 1.86 µg/m^3. A distinct bimodal distribution of BC concentrations was observed across all seasons during rush-hour traffic, with a morning peak between 10:00-11:00 and an evening peak around 20:00-21:00. The lowest concentrations consistently occurred between 05:00-06:00. Seasonal variations showed the highest concentrations in winter and autumn, followed by spring, with the lowest levels observed during summer, consistent with trends in neighbouring countries. Fossil fuel combustion was the dominant source of BC at Sofia site across all seasons, accounting for about 91% of total BC, while biomass burning contributed around 9%.The percentage contribution of biomass burning to BC (BB%) - from sources such as residential heating, agricultural burning, and wildfires - was most pronounced in winter. BB% was significantly higher during weekends across all seasons, particularly in winter, likely due to reduced traffic emissions and increased residential wood burning.

BC concentrations showed an inverse relationship with both wind speed and temperature. Higher BC levels were associated with calm and colder conditions, consistent with typical urban pollution dynamics, where stagnant air favours pollutant accumulation, while higher wind speeds and temperatures promote dispersion and dilution.

A limitation of this study is the lack of information on site-specific MAC values. As recommended by Savadkoohi et al. (2023), obtaining a site-specific MAC through comparisons with co-located elemental carbon (EC) measurements is important. Such a study in Sofia is feasible and may be part of a future project.

Nevertheless, it is important to note that this study provides valuable insights into air quality in the largest city of Bulgaria and contributes to filling the gap in black carbon (BC) datasets in Southeastern Europe.

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