Concentration and source allocation of black carbon by AE-33 model in urban area of Shenzhen, southern China

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Abstract

Purpose In the urban region of Shenzhen, the changes in the concentration of Black carbon (BC) have been evaluated throughout the dry season, and apportioned the BC sources, including in the form of fossil fuel (e.g., vehicle emissions) and biomass fuel (e.g., industrial emissions).

Methods The new seven-channel aethalometer model (AE-33), PM$_{2.5}$, and meteorological data were collected in the dry season (October–May) from 2019 to 2020, to quantify BC emissions in urban Shenzhen. Explored the source allocation of BC based on Potential source contribution function (PSCF) and concentration-weighted trajectory (CWT) model.

Results We revealed that the mean BC concentration was 2672 ± 1506 ng/m$^3$ in the dry season, with values of 4062 ± 1182 ng/m$^3$, 2519 ± 1568 ng/m$^3$, and 1900 ± 776 ng/m$^3$ in autumn, winter, and spring, respectively. Additionally, we found that fossil fuels have higher contributions to BC concentrations (86.3% to 86.8% in autumn and spring) in the dry season than biomass fuels (16% to 20% in autumn, spring and winter), which is different from Beijing, Nanjing and other large economic zones in China. The diurnal variation in BC and the contribution of fossil fuels indicate that there is a significantly greater increase in BC during peak traffic hours in urban Shenzhen than in other cities. Finally, meteorological parameters and PM$_{2.5}$ data provided supporting evidence that BC is sourced mainly from local vehicle emissions and industry-related combustion in the western and northeastern/southeastern parts of the study area.

Conclusion This study showed that the concentration of BC is lower than other regions, and the source allocation is mainly local fossil fuels (vehicle emission, etc.).

Keywords Black carbon · Shenzhen · Dry season · Vehicle emission · Transport

Introduction

Black carbon (BC) is the main pollutant resulting from the incomplete combustion of biomass and fossil fuels and emitted into the atmosphere. Moreover, BC is the most incombustible and light-absorbing component of carbonaceous aerosols and the core structural component of PM$_{2.5}$ [24, 26, 33]. In recent decades, because BC can penetrate human lungs and bronchi, many studies have reported the relationship between cardiovascular and respiratory diseases and BC [23, 27], and BC has garnered increasing research attention. In addition to its light absorption property, which reduces atmospheric visibility and deteriorates local and regional air quality, BC has a great impact on global warming due to its unique optical and radiation characteristics [12, 22, 25, 41]. To decrease the effect of BC on human health and the climate, it is necessary to identify and quantify the contributions of different sources of BC, which can facilitate the control of BC emissions [2].
Carbon isotope and aethalometer models provide mainly quantitative research on BC source allocation [20, 36]. The isotope method can quantify different sources of BC (e.g., fossil sources and biomass combustion) directly but can only be applied to filtered samples, so the temporal resolution of this method is limited. Compared to isotopes, aethalometer models (AE-31 and AE-33) are suitable for research on BC source allocation in the long term due to their high temporal resolution, and these models have been widely used [28]. The aethalometer model is based on the wavelength-dependent difference in the absorption coefficient and assumes that the absorption of biomass emissions and fossil fuel follows the power law $b_{abs} (\lambda) \sim \lambda^{-\alpha}$, where $\lambda$ and $\alpha$ represent the wavelength and absorption Ångström exponents, respectively [1, 7, 40]. Therefore, considering the air pollution in Shenzhen, we applied the aethalometer model to track the sources of BC in Shenzhen. In general, aethalometer models have been effective in some regions (e.g., India, Beijing, and Nanjing) [6, 20], but there is no research to date in Shenzhen using AE-33 (the updated version of AE-31).

Jing used the aethalometer model to apportion the contributions of fossil fuel (traffic) and solid fuel sources to BC, and liquid fuel had higher contributions (70% to 84% in autumn and winter) to BC concentration than did solid fuel (e.g., coal and biomass, ranging from 16% to 30% in winter-autumn) in Nanjing [19]. Liu reported that during haze days, solid fuel sources play an important role in BC contributions, whereas on haze-free days in Beijing, liquid fuel sources contribute more to BC [21]. In western India, previous studies showed that the contribution of fossil fuel to BC was 2–4 times higher than that from wood burning according to an aethalometer model. Hua et al. [17] showed that fossil fuel combustion accounted for 82.5%, 82.9%, and 85.5% of the total BC in sampling sites in urban, suburban, and rural Nanjing, respectively (13, 17, 21).

The aethalometer model has generally been effective for identifying the contributions of fossil fuel and biomass burning to BC in different regions [7, 14]. Most research has used the AE-31 aethalometer model. AE-33, a new BC measurement method, is an upgraded version based on AE-31 [3, 28]. However, few studies have used this new BC measurement approach. In addition, Shenzhen has more vehicle ownership and a greater population than in many other studied areas. In 2015, a PM$_{2.5}$ source analysis by the Ministry of Environmental Protection showed that the primary source of PM$_{2.5}$ in Shenzhen was motor vehicles [9]. Therefore, BC contributions in urban areas are extremely important, and a high temporal resolution is necessary for research on BC source allocation on a long-term scale. However, there is no research on BC from different sources (e.g., fossil fuel and biomass burning) employing the aethalometer model, except for a report by Cheng et al., who used an AE-31 instrument in Shenzhen in 2018 [3, 7]. Due to its efficiency, AE-33 can measure BC concentrations more accurately than AE-31 and should therefore be applied to various regions.

In the present research, we explored the application technology of the AE-33 aethalometer model in Shenzhen. The region is adjacent to Hong Kong and is an important economic area of the Greater Bay Area (Guangdong-Hong Kong-Macao, GBA), where pollution is mainly determined by vehicle usage and variations in meteorological conditions. Aethalometer, PM$_{2.5}$ and meteorological data were collected in the dry season (October–May) from 2019 to 2020 to quantify BC emissions in urban Shenzhen. We apportioned the BC sources in urban areas, including in the form of fossil fuel (e.g., vehicle emissions) and biomass fuel (e.g., industrial emissions), and explored the characteristics of BC concentrations and different sources during the dry seasons in Shenzhen based on the aethalometer model.

Methodology and data

Study area

Shenzhen is on the coast in Guangdong Province (22°33’N, 114°06’E) in the GBA near Hong Kong, China (Fig. 1). The city has 13.66 million residents, and more than 3.5 million vehicles were used in 2019. Because of the high output of industrial activities, the rapidly increasing population, and the soaring number of transportation vehicles, Shenzhen is the most advanced zone of economic demonstration in China. Shenzhen has been experiencing an increase in the pollution level of BC due to its rapid economic development in recent decades. BC concentrations have been measured in Shenzhen, and a previous study demonstrated that the mean concentrations of BC were $1120 \pm 90$ ng/m$^3$ and $2580 \pm 200$ ng/m$^3$ in suburban and urban areas, respectively [7]. The stable stratification and low-speed winds that accompany stable high pressure and the degeneration of high-pressure ridges make it difficult for air pollutants to diffuse, which is one of the main causes of regional air pollution. Negative and positive temperature changes before the front, boundary layer inversion, a wide diurnal temperature range and the low height of the mixed layer are all conducive to the generation of haze weather. Local circulation phenomena such as sea and land breezes, valley breezes, and heat island circulation also have significant impacts on the atmospheric boundary in Shenzhen. Related research has shown that PM$_{2.5}$ is an important pollutant. Thus, monitoring the concentration of BC, which is a core structural component of PM$_{2.5}$, and identifying its source allocations can help to reduce BC emissions and improve public health and the atmospheric environment in the GBA.
Methodology

Sampling

An aethalometer (Model AE-33, Magee Scientific, USA) was used to measure the BC concentration from October 2019 to May 2020 at the College of Chemistry and Environmental Engineering of Shenzhen University (22.35°N, 113.59°E), as shown in Fig. 1. The sampling site is located 60 m above the ground on the rooftop of a building at Shenzhen University, and the air inlet is installed at 65 m. Related research used one sample point at this height to represent the BC concentration in city regions, demonstrating that this method can be used to study the trend of BC concentration in urban areas. Therefore, one sample point at each sampling site was used in our research [7, 20, 40]. If special weather conditions were present, such as smoke generated by a nearby fire, we excluded the data. The western/northwestern sampling site is in a zone of industry with factories (Baoan, Guangming and Longhua districts), including manufacturing facilities for electronics, chemicals, paper products, plastics and hardware, clothing, machinery and electrical machinery. In addition, there are many roads and constantly operating construction companies around the sampling site. The horizontal and vertical distances between the construction area and site are approximately 500 m and 110 m, respectively. The sampling site is located in a mixed area of residential and industrial regions, representing a typical urban area in Shenzhen.

The AE-33 aethalometer continuously collects aerosol particles by passing the aerosol-laden air stream through points on the filter belt. Analyses were performed at seven optical wavelengths (λ = 370, 470, 520, 590, 660, 880 and 950 nm), ranging from near infrared to near ultraviolet [28]. We used the variations in optical attenuation and a mass absorption cross section (MAC) of 7.77 m²/g at 880 nm to calculate the BC mass concentration in the selected time interval [15]. PM$_{2.5}$ data were measured at the Huaqiaocheng station. The horizontal distance between the Huaqiaocheng station and the site at Shenzhen University is approximately 8 km. Both are state-controlled air sampling stations in Shenzhen and are affected by almost the same emission sources of BC and other pollutants, so they can represent typical pollutants in the local region. Therefore, we believe that the impacts of using these two sites on the methodology are small and that the data from both sites can be used in research. The wind direction and speed were collected from the same locations as the BC concentration measurements and were input as meteorological parameters into the National Centers for Environmental Prediction-National Center for Atmospheric Research (NCEP/NCAR) Gridded Analysis and Display System (GrADS).
Aethalometer model

Measurement principle

The measurement of BC concentration is based on light absorption, which is measured on a filter loaded with aerosols [18]. Field validation was performed to ensure that the instrument met the study requirements. AE-33 is an upgraded version of AE-31, and it provides a measurement of BC concentration that has been recognized in other regions, such as Beijing, Shanghai, and Nanjing. Through a filter on which aerosols are continuously collected, the attenuation (ATN) of a transmitted light beam is measured from the near infrared to the near ultraviolet wavelengths [41]. During data processing, based on the running state value recorded during the sampling of the BC meter, obvious abnormal values (such as negative values) were eliminated. The processed effective data minutes accounted for more than 90% of the original data, ensuring the scientific rigor and rationality of the data. To decrease the effect of BC on human health and the climate, it is necessary to explore the sources of BC and quantify the contributions of different sources to BC, which can provide essential information to help control BC emissions [2].

\[
ATN = 100 \times \ln \left( \frac{I_0}{I} \right) \tag{1}
\]

where \( I_0 \) is the reference signal and \( I \) is the measured signal. Based on ATN, the light absorption coefficient of BC \( (b_{abs}) \) at distinct wavelengths can be calculated by AE-33 as follows:

\[
b_{abs} = \frac{b_{ATN}}{C} \tag{3}
\]

where \( b_{ATN}, F, S, C, t \) represent the coefficient of optical attenuation, flow rate, spot area, multiple scattering parameter and measured time, respectively. The absorption of non-BC particles decreases as the wavelength increases, and the rate of decrease is faster than that of BC. Therefore, according to this principle, the absorption percentage of BC in the long-wavelength bands will be greater than that in the shorter-wavelength bands. The absorption of other aerosols (e.g., mineral dust or brown carbon) is not sensitive at 880 nm [26]. According to this principle, the instrument has been widely used in other areas. The data obtained from channel 6 are the authoritative standard for reporting BC concentrations. Therefore, based on the MAC and absorption coefficient, the BC concentration can be calculated at 880 nm. For more detailed information, see Drinovec et al. [11].

Model

In the current study, the source allocation of BC emissions was based on the model of Sandradewi et al. [28], which showed that fossil fuel burning and biomass burning together form the optical absorption coefficient [28]. The model is based on the wavelength-dependent difference in the absorption coefficient and assumes that the absorption of biomass emissions and fossil fuel follows the power law \( b_{abs}(\lambda) \sim \lambda^{-\alpha} \), where \( \lambda \) and \( \alpha \) represent the wavelength and absorption Ångström exponents, respectively. The different \( b_{abs}(\lambda) \) values at 370 nm and 880 nm were used to calculate the \( \alpha \) value. However, apart from BC, the absorption of other aerosols is not sensitive at 880 nm. The model assumes that the total light absorption at 880 nm mainly comes from the BC in fossil fuels (i.e., traffic sources) and biomass fuels (i.e., coal and biomass combustion). In addition, previous studies have shown that BC comes from the combustion of solid fuels and liquid fuels in China, and the Shenzhen sampling site has obvious vehicle emission sources and industrial emissions. Therefore,

\[
b_{abs}(BC) = b_{abs}BC_{fossil} + b_{abs}BC_{biomass} \tag{4}
\]

\[
b_{abs}(370nm)_{fossil} = \left( \frac{370}{880} \right)^{-\alpha_{fossil}} \tag{5}
\]

\[
b_{abs}(880nm)_{fossil} = \left( \frac{370}{880} \right)^{-\alpha_{fossil}} \tag{6}
\]

where \( \alpha_{biomass} \) and \( \alpha_{fossil} \) represent the biomass fuel-derived BC and light-absorption Ångström exponent for fossil fuel-derived BC, respectively. Therefore, different sources of BC can be identified based on the coefficients of light absorption and MAC (7.77 m² g⁻¹) [11, 15],

\[
BC_{source} = \frac{b_{abs}(880nm)_{source}}{MAC \times (880nm)} \tag{7}
\]

where BC source represents the source of BC, i.e., fossil fuel or biomass fuel. The contribution of fossil fuels (C) is

\[
C = \frac{b_{abs}(880nm)_{fossil}}{b_{abs}(880nm)} = \frac{BC_{fossil}}{BC} \tag{8}
\]

Potential source contribution function (PSCF)

The potential source contribution function (PSCF) value of grid cells in the research domain is determined by calculating the
endpoints of the trajectory segment ending in each cell [4]. The number of endpoints that fall into the \(ij\)-th cell is called \(n_{ij}\). The number of endpoints in the same cell with an arrival time to the sampling point corresponding to a BC concentration higher than an arbitrarily set standard is defined as \(m_{ij}\). Then, the PSCF value of the \(ij\)-th pixel is defined as

\[
PSCF_{ij} = \frac{m_{ij}}{n_{ij}}
\]

(9)

The PSCF is defined as a conditional probability, in which the concentration of a given analyte is greater than a standard level, and in the process of transport to the receptor site through the \(ij\)-th cell, the PSCF is related to the passage of air packets [8]. In other words, if a cell with high values in PSCF are related to the air packet reaching the receptor site, the analyte concentration of these packets is higher than the standard level. This condition characterizes regions where the component has a “high potential” contribution.

**Concentration-weighted trajectory (CWT)**

In the concentration-weighted trajectory (CWT) method [16, 20, 29], the mean concentrations of samples can be used to calculate the weighted concentration of each grid cell with the relevant trajectory crossing the grid cell. The more traversed trajectories there are and the longer the dwell time is, the more likely grid cells are to contribute to the observed properties of aerosols. The region of interest is divided into a \(0.5^\circ \times 0.5^\circ\) grid, and a weighted value is assigned to each grid point according to the value observed at the receptor site when the relevant trajectory crosses the grid point \((i, j)\). The sample data are obtained by averaging as follows:

\[
C_{ij} = \frac{1}{\sum_{l=1}^{M} r_{ijl}} \sum_{l=1}^{M} C_{i} r_{ijl}
\]

(10)

where \(C_{ij}\) is the average weighted concentration of the \(ij\)-th pixel, \(l\) is the index of the trajectory, \(M\) is the total number of trajectories, \(C_{i}\) is the concentration observed when trajectory \(l\) arrives, and \(r_{ijl}\) is the time the trajectory spent in the \(ij\)-th pixel. A higher value of \(C_{ij}\) means that the air packet traveling through the \(ij\)-th pixel is, on average, correlated with a high concentration at the receptor. The abovementioned arbitrary weighting function is also used in CWT analysis to reduce the influence of smaller values of \(n_{ij}\).

**Results and discussion**

**BC concentration**

Both hourly and daily average BC concentrations were calculated from 1 October 2019 to 31 May 2020 in Shenzhen and are shown in Fig. 2. This region has annual wet (May or June to September or October) and dry seasons (September or October to April or May) [7]. According to previous studies, April–September is the wet season and October–January is the dry season in Guangdong Province, and February–March is the transition period from the dry season to the wet season. The climate in Shenzhen is slightly different. The data we obtained on the weather conditions from October 2019 to May 2020 indicated that this period constitutes the dry season. There is a large amount of rain in the wet season, the deposition of pollutants is severe, and the BC concentration is much lower than that in the dry season [34]. Therefore, we focused on the analysis of variations in BC concentration and its potential sources over Shenzhen in the dry season (Fig. 2a and b) from 1 October 2019 to 31 May 2020.

The hourly BC concentration ranged from 268 to 16,293 ng/m³, and the average concentration and standard deviation were 2672 ng/m³ and 1506 ng/m³ in the dry season, respectively. The mean monthly concentration of BC in Fig. 2 shows the highest value (3273 ± 1569 ng/m³) and the lowest value (548 ± 325 ng/m³) in November and May, respectively. The mean BC concentration in the dry season was in the following order: autumn (4062 ± 1182 ng/m³) > winter (2519 ± 1568 ng/m³) > spring (1900 ± 776 ng/m³). The average concentration in autumn was greater than that in winter. The main reason is that during the winter of 2019, due to the impact of COVID-19, the number of vehicles in the urban area was greatly reduced, so the concentration in winter was lower than that in autumn. Compared with other seasons, the BC concentration in autumn was approximately doubled, which may be due to the lower mixing height resulting in a more stable atmosphere. Moreover, BC concentrations exhibited the same seasonal variations in 2018, with the lowest value in spring in Shenzhen [7]. However, the results of this experiment show that although the maximum concentration occurred in winter, the autumn average value was greater than the winter average value. This result may be related to the occurrence of COVID-19 in 2020, resulting in a reduction in vehicle emissions in Shenzhen due to the BC concentration being significantly lower than that in previous years from January to February. Nevertheless, due to the atmospheric boundary layer and wind speed, the highest value still appeared in January.

The weather associated with pollution in Shenzhen can be divided into two types: continental high-pressure type and tropical cyclone type. The continental high-pressure type is the most common, accounting for close to 90% of pollution-related weather. This weather type mainly occurs in autumn and winter. Rainfall in autumn and winter is significantly reduced, cold air activities are frequent, and northerly winds prevail. The northerly airflow transports particulate pollutants from north to south, and the frequency of calm winds...
increases in winter. Winter is the weakest wind season in Shenzhen. The frequency of inversion weather is high, and the atmospheric diffusion conditions are poor, causing air pollutants to accumulate for a long time.

Compared with other seasons, the anthropogenic BC emissions from fossil fuels in winter in China are relatively high [14]. BC concentrations are higher in winter because the frequency of the temperature inversion layer is high, which is not conducive to the diffusion of pollutants [37].

To further compare the BC concentrations at other stations with those in Shenzhen, Table S3 shows the BC concentrations observed in China (major pollution areas). In general, the concentration of BC in urban areas is higher than that in suburban and mountainous areas. In addition, the BC level in Beijing-Tianjin-Hebei may be higher than the BC level in the Pearl River and Yangtze River Delta. Studies have shown that China’s BC mainly comes from coal combustion, automobile exhaust emissions and biomass combustion [30]. In the region of Beijing, the BC concentration surpasses 10,000 ng/m³, and the burning of fossil fuels is extensive [13, 35], with a value approximately 4 times that in urban Guangzhou. Although the annual BC concentration is relatively low in the suburbs of this region, there are large differences in pollution values in all seasons and daily. Therefore, it is necessary to analyze the sources of BC in detail, and we found that traffic emissions are a main source of BC in Shenzhen [7, 40].

Source allocation of BC by the aethalometer model

We used the aethalometer model to quantify the contributions of BC sources in urban Shenzhen in the dry season. Previous research has consistently shown that BC mainly comes from fossil fuel combustion (from vehicles) when $\alpha$ is close to 1, whereas when $\alpha$ is approximately 2.0, BC mainly comes from biomass burning [28].

In the aethalometer model, fossil combustion and biomass burning are the two main sources of BC, which is consistent with findings in other countries [41]. Research in Shenzhen based on the AE-31 model indicated that the daily variation in BC concentration fluctuates over a large range and has an obvious double-peak structure. The two peaks appear at approximately 08:00 in the morning and 19:00–21:00 in
the evening, which correspond to the morning and evening
peaks of vehicle emission sources in the urban area. How-
ever, the different efficiencies and types of combustion can
determine the $\alpha$ value. To determine the $\alpha$ values of fossil
fuels and biomass fuels in Shenzhen, we used the approach
of Zotter et al., in which a linear fitting relationship between
the annual $\alpha$ and C values was used [28].

Based on the research of Sandradewi et al., for the par-
ticulate matter emitted by fossil fuel combustion, $\alpha$ is usu-
ally close to 1, and the $\alpha$ value of biomass combustion is
approximately 1.8–1.9. In addition, Peter Zotter used the $^{14}$C
tracer method for comparison with the results of Sandradewi
to accurately determine the specific $\alpha$ values correspond-
ing to fossil fuel combustion and biomass combustion in
Switzerland [41]. When the best fit was reached ($r=0.81$),
coal combustion and biomass combustion corresponded
to $\alpha$ values of 0.9 and 1.68, respectively. These results are
more reliable than previous results because of the use of
isotope tracing. The daily C and $\alpha$ values in the dry season
showed a high negative correlation in Shenzhen ($R^2=0.85$).
Thus, 0.9 and 1.68 were chosen to correspond to fossil fuel
combustion and biomass combustion, respectively, in the
present study.

The average monthly values of $\alpha$ and C are shown in
Fig. 3a and b. There was a large change in $\alpha$ and C in each
month, which indicated that the source intensity of BC
emissions varied, with daily values of $\alpha$ and C ranging from
0.8 to 1 and 83% to 89%, respectively. We calculated the
mean BC concentrations in different months and seasons
and then determined that the hourly average $\alpha$ varied from
0.188 to 1.48 in the dry season. The $\alpha$ values were as follows
in the dry season: spring (0.868) > autumn (0.838) > winter
(0.811). The C values were arranged from high to low in
the dry season as follows: spring (0.867), autumn (0.861),
and winter (0.795). The results indicated that the BC mainly
comes from fossil fuel sources (vehicle emissions) in the
dry season, especially during autumn and spring. Previous
studies have reached the same conclusion: the BC at all sites
originated from the burning of fossil fuels, most of which is
attributable to vehicle exhaust emissions. Therefore, reduc-
ing vehicle emission sources is very important for human
health, and a suitable plan must be established to accomplish
this reduction.

The pollution in Shenzhen is formed under the combined
influence of the large-scale circulation background and the
local sea-land breeze circulation. In spring, it is mainly a
weak circulation field controlled by a high-pressure ridge of
degeneration, which provides conditions for the formation
of sea-land breeze circulation. In autumn, the circulation
is dominated by a weak high-pressure ridge under the con-
trol of the subtropical high. The convergence of dominant
winds and land-sea winds forms along the coast, causing
short-term local pollution. With the destruction of circula-
tion and the disappearance of the inversion layer, the pollu-
tion disappears. Winter is mainly under the influence of the
weak circulation field controlled by the cold front or weak
pressure ridge.

Similar to other studies, we found that the BC concen-
tration emitted by vehicles in Shenzhen is very high, espe-
cially in autumn and spring. However, due to COVID-19,
less travel and work occurred during the Spring Festival
holiday, and the BC concentration in the winter of the study
year was lower than that in previous years. However, the
government still must take preventive measures to reduce
the concentrations of BC in autumn, winter and spring dur-
ing the dry season.

The contributions of BC pollution emission sources at
the site are relatively stable, and the increase or reduction
of a certain emission source has a minor impact on BC. The monthly $\alpha$ value fluctuates by a value of approximately 1, which shows that the main source of BC pollution is mostly from the burning of fossil fuels. A large portion of the pollution at the site is due to the increased BC concentration caused by vehicle exhaust emissions, which is consistent with previous research.

Beegum et al. [5] reported that the diurnal variation in BC is typically used for mainland sites [5]. In general, the daily BC concentration in autumn-winter was higher than that in spring (Fig. S1), and the variation in BC was mainly affected by different sources of emissions and meteorological changes [21]. The variations in $C$, $\alpha$ and BC in the dry season are shown in Fig. 4a–d and 5 and Fig. S1. The horizontal axis of Fig. 4 is 24 h a day, from 00 to 23. BC has been regarded as the core structural component of PM$_{2.5}$, impacts from boundary layer mixing and anthropogenic emission same as PM$_{2.5}$. In Shenzhen, concentration of PM$_{2.5}$ decreases with the increase in height, and the correlation of the concentration of PM$_{2.5}$ at different heights is poor. The boundary layer causes the correlation between the low and high pollutants to become worse at night. PM$_{2.5}$ concentration at 60 m/70 m is higher, which all reflects the significant impact of ground emissions. At 210 m and 325 m, the diurnal characteristics is “high during the day and low during the night”, reflecting the process of mixing and spreading from the ground to the high altitude during the day [32]. The daily variation in BC concentration fluctuates over a large range, with an obvious double-peak structure. The two peaks appear at approximately 08:00–11:00 in the morning and 19:00–21:00 in the evening. The morning and evening peaks of the sources correspond. The valleys appear at approximately 13:00 at midday and 04:00 at night. However, due to the Spring Festival holiday, there were fewer vehicle emissions in the winter period. The time of the first valley is related to the uplift of the boundary layer. In the dry season, traffic sources for BC were the main factor, contributing more than 85% to the total BC on average. The diurnal variation characteristics of BC are determined by the combined influence of diurnal changes in meteorological conditions and diurnal variations in anthropogenic emissions, which

![Fig. 4](image_url)
can reflect atmospheric boundary layer conditions and diurnal variations in anthropogenic activities to a certain extent [31].

The high BC concentrations at the above times of day may be due to meteorological conditions and increased emissions from traffic (there is a main road with substantial vehicle emissions around the station). Based on a comparison with the observed value in Shenzhen in 2012 (5700 ng/m$^3$) [7], the BC concentration in the urban area of Shenzhen has decreased significantly; this change indicates that the air quality in the urban area of Shenzhen in recent years has improved, probably because the government promoted the use of electric buses.

**Supporting evidence for the results of BC sources**

**PSCF analysis in the dry season**

Relevant studies have shown that solar activity also has an impact on the monsoon. In years of high solar activity, in most Asian summer monsoon regions, there is a consistent increase in the near-surface southerly wind, which affects atmospheric convection and precipitation [10]. However, our study focuses on the dry season in Shenzhen, which does not include summer, and we did not investigate the effects of variations in solar activity on the transport pathways of BC.

The changes in BC concentration in the three seasons may be affected by the transportation processes in different regions. Figure 6 shows the results of PSCF over the three seasons. In addition to local pollution sources, we analyzed the impact of long-distance transportation. The northeastern sampling site has air masses, and the source areas with the highest potential contributions in the three seasons are the central and eastern parts of Guangdong, southwestern Jiangxi, western Fujian Province, and Hong Kong. From Fig. 6a–d, we find that the source areas with high contribution potential in autumn are larger than those in winter and spring. In autumn-winter, the areas of high potential sources (PSCF>0.7) are all in Guangdong and Jiangxi, Fujian Province. In spring, the regions of sources with high potential
(PSCF>0.7) are mainly in the direction of Hong Kong, and pollutants are discharged from the eastern Taiwan Strait and nearby islands.

Throughout the dry season, the pollution sources mainly come from Guangdong, Jiangxi and Fujian Provinces in the northeast and Hong Kong and western Shenzhen in the southeast, and the main airflow is formed by the superposition of pollutants discharged by surrounding cities and local emission sources. Therefore, BC from the southeast and northeast regions (mainly Guangdong and Jiangxi Provinces in all seasons) contributed substantially to the BC concentration.

BC is an important component of PM$_{2.5}$; we analyzed the potential sources of PM$_{2.5}$ in the dry season of 2019–2020 (Fig. 7). The maximum value (PSCF>0.4) still came from Jiangxi, Guangdong and Fujian, where fixed pollution sources in Jiangxi stably transport pollutants to the sampling point. Local pollution sources are also located in high-value areas in the Baoan and Longhua districts in Shenzhen. From the analysis of the relationship between BC and PM$_{2.5}$ (presented in section 3.3), the high potential sources in the dry season are mainly from local pollution and remote contamination resulting from long-distance transportation.

**Centers of high PM$_{2.5}$ concentration in Shenzhen**

BC is mainly derived from the incomplete combustion of carbon-containing fuels and is an important component of PM$_{2.5}$. Many studies have monitored the concentrations of PM$_{2.5}$ and BC at the same time to measure local pollutants [8]. Therefore, we also used the PM$_{2.5}$ concentrations from 2016 to 2018 in Shenzhen to predict the key polluted regions to supplement the results of the analysis of potential local sources of BC. PM$_{2.5}$ concentrations with high potential were calculated by the k-means cluster method, in which the migration of the center of gravity of PM$_{2.5}$ over Shenzhen was analyzed (Fig. 8). Two centers are clustered by the centers of gravity in each map of PM$_{2.5}$, which indicates that the regions of sources with high pollution potential are mainly two regions over Shenzhen. As shown in Fig. 8, the
regions of high pollution potential are stable, and 75% of
the points fall within the cluster range. There are two
high-value centers of PM$_{2.5}$ concentration in Shenzhen according
to the clusters. One source region is located at the junction
of the Yantian and Pingshan districts, where the PM$_{2.5}$ value
ranges from 52 to 55 μg/m$^3$, and another source region is
located at the junction of the Longhua, Baoan, Nanshan and
Guangming districts, where the PM$_{2.5}$ value ranges from 63
to 69 μg/m$^3$. The high potential PM$_{2.5}$ sources mainly come
from Pingshan and Baoan districts over Shenzhen, which
have high annual and interannual values. Shenzhen is led by
the wind direction of the northeast in the dry season, where
pollutants (PM$_{2.5}$ and BC) are mainly concentrated in the
regions of Longhua, Baoan, Yantian and Pingshan.

In addition to weather conditions, marine and terrestrial
aerosols originate from atmospheric pollution along urban
coasts, the sources of which come from complex chemical
and physical activities. The PM$_{2.5}$ high-value area is consist-
ent with the BC high-potential source regions, again indicating
that the sources of BC in Shenzhen are mainly from local
pollutants from the west and southeast of the sampling site.

**Analysis of high BC concentrations in the dry season**

In this study, we also analyzed the correspondence between
airflow based on meteorological data and BC concentration.
Figure 9b shows the airflow trajectory clustering results for
the station for 24 h and the corresponding BC concentration
distributions of the clusters in different directions, which
represent the clustering results for the dry season. The BC
concentration at the site is mainly affected by the pollut-
ants emitted by local emission sources in the dry season.
The contributions of airflow sources from different direc-
tions of the station follow the order of southeast airflow
(48%) > northeast airflow (29.87%) > south airflow (22.13%).
The corresponding BC averages in different directions in
the dry season are arranged in descending order as follows:
southeast airflow (4610 ± 221 ng/m$^3$) > northeast airflow
(4100 ± 236 ng/m$^3$) > south airflow (3460 ± 156 ng/m$^3$). The
airflow with a BC concentration greater than 3000 ng/m$^3$ and
PM$_{2.5}$ concentration greater than 40 μg/m$^3$ mainly comes
from Jiangxi in the northeast direction, and it accounts for
approximately 30% of the total airflow.
Fig. 8 Migration of the center of gravity of PM$_{2.5}$ concentrations in Shenzhen in dry seasons

Fig. 9 A show the CWT air mass back-trajectories at 500 m, and b corresponds to the BC level in the dry season
The trajectories with a BC concentration greater than 1000 ng/m³ in months other than May accounted for 98.9% (October 2019), 95.6% (November 2019), 93.2% (December 2019), 85% (January 2020), 66.2% (February 2020), 83.5% (March 2020), and 85.5% (April 2020); only 56% was observed in May (Fig. S2). The BC concentrations corresponding to the above pollution trajectories are 3795.94 ng/m³ (October 2019), 4260.8 ng/m³ (November 2019), 4263.57 ng/m³ (December 2019), 2076.25 ng/m³ (January 2020), 1524.13 ng/m³ (February 2020), 1954.29 ng/m³ (March 2020), 2346.57 ng/m³ (April 2020), and 2129.77 ng/m³ (May 2020) (Fig. S2).

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**Comparison with previous studies**

Radiocarbon analysis is an important tool to unambiguously distinguish fossil and non-fossil sources of carbonaceous particles. All fossil fuel emissions, such as petroleum and coal combustion, are 14C free, whereas non-fossil emissions (e.g., biogenic emissions, biomass burning, cooking) contain 14C [38]. However, radiocarbon analysis can only be applied to filtered samples, so the time resolution is limited. Briefly, the instrument used is composed of four main parts: a hood for gathering emissions from stoves and clean air around the area; a long, curved pipe immersed in water for cooling flue gas to ambient temperature; an end pump for drawing the flue gas through the pipe system at a flow rate of approximately 1 m³/min and blowing the exhaust outside the sampling room; and a branched pipe upstream the end pump for ducting a portion of exhaust to two samplers [39]. Compared to isotope tracing, the aethalometer model (AE-31 and AE-33) is suitable for research on BC source allocation in the long term due to its high temporal resolution (specific description in section 2.3.2) according to our research in the Shenzhen region. Therefore, we used the aethalometer model in the present research.

For comparison with the BC concentration measured by the aethalometer model (AE-31) in 2014, we used AE-33 (upgraded version of AE-31) to measure the BC concentration, and the results showed that the concentration of BC in urban Shenzhen declined from 2019 to 2020. In 2014, the concentration in Shenzhen in the dry season was approximately 3260 ng/m³, and the concentration obtained in our research was 2672 ng/m³; this decrease is credited to the government’s effective work related to promoting electric public transportation [7]. On the basis of the aethalometer model, the different sources of BC in Shenzhen are mainly vehicle emissions, and the BC concentration map is consistent with the values during the peak rush hours (08:00–09:00 and 19:00–21:00), except during the Spring Festival. In general, the BC concentration distribution is also related to the coastal geographic location and meteorological conditions of Shenzhen. Relevant studies in Beijing and Nanjing have shown that BC has additional sources of biomass combustion, especially in autumn and winter, which is related to the local atmospheric boundary layer and industrial structure [13, 20]. The Ångström exponents (α) of Beijing and Nanjing are also very different from that of Shenzhen. Shenzhen’s α is approximately 1, while the seasonal average α in Beijing and Nanjing can reach approximately 1.3 or even 1.6, which again illustrates the different sources of BC in different cities [21]. The BC concentrations of urban areas in Beijing and Nanjing are much higher than those in Shenzhen. The average BC concentration in the Beijing urban area is 11,100 ng/m³, and the BC concentration in the Nanjing urban area is 4157 ng/m³ [35, 40]. In summer, the concentrations in Beijing and Nanjing are approximately 2–3 times higher than those in Shenzhen.

Although we only measured the BC concentration in the dry season, the basic concentration variations and source analysis of BC in Shenzhen could still be analyzed by the aethalometer model. Overall, the model is superior in performance to previous models, and this is the first time an AE-33 instrument has been used in Shenzhen to measure BC concentrations and PSCF potential sources; this study can thus lay the foundation for local pollutant concentration measurements and source tracking.

**Conclusions**

The source allocation of BC in Shenzhen in the dry seasons from 2019 to 2020 was performed on the basis of an aethalometer model (AE-33). In general, the daily BC
concentrations in autumn-winter are higher than those in spring, which fluctuate over a large range with an obvious double-peak structure. Moreover, meteorological and source emission variations are the main reasons for the differences in BC concentrations. The sources are quantified based on the BC from fossil fuel (e.g., traffic emissions) and biomass burning sources (e.g., industry). Traffic sources for BC are the main factor, contributing more than 86% of the total BC on average. The diurnal BC concentration shows an increasing trend during peak hours. Traffic emissions play an important diurnal role in BC based on the variations in C and α, and the C parameters maintain high values during peak hours throughout the day. In autumn, spring and December, fossil fuel sources (mainly vehicles) show an increased contribution to the BC concentration. The PM$_{2.5}$ results in Shenzhen are consistent with the BC source results based on the aethalometer model in this study. In addition, long-distance transportation from the southeast and northeast of the sampling site was also an important factor in BC concentration in different seasons. High BC levels mainly come from the west, northeast, and southeast in Shenzhen, and transport from these directions is the main source of BC in addition to local vehicle emissions and long-range transport from industrial areas in other regions. A high contribution to BC also comes from the southeastern direction of Hong Kong in the dry season. The CWT method suggests that the high BC in the dry season is caused by local activities in the western and northeastern regions of Guangdong and Jiangxi Provinces.

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Availability of data and materials All data generated or analyzed during this study are included in this published article and its supplementary information files.

Declarations There is no conflict of interest in the submission of this manuscript, and the manuscript has been approved by all authors for publication. All of the authors declare that the work described is original research that has not been published previously and is not under consideration for publication elsewhere, in whole or in part. All the authors listed have approved the enclosed manuscript. We further confirm that the order of authors listed in the manuscript has been approved by all of us. Wenqian Chen performed the experiment and wrote the manuscript. Xiaoyi Cao contributed meaningfully to the analysis and manuscript preparation, Haofan Ran performed the data analyses, Ting Chen and Bohan Yang helped perform the analysis with constructive discussions, and Xuan Zheng performed the experiment and manuscript preparation. This work is supported by Guangdong of China Regional Youth Foundation (2019A1515110692), Chinese Postdoctoral Science Foundation (2020 M682856) and National Natural Science Foundation of China (51978404). We are especially grateful to the editors and anonymous reviewers for appraising our manuscript and for their instructive comments.

References

1. Ahmed TD, Khan VA, A. J. Long term trends in black carbon concentrations in the northeastern United States. Atmos Res. 2014;137:49–57.
2. Aljefri L. Development of a method for classification of hospitals based on results of the diagnosis-related groups and the principle of case-mix index. Eastern Mediterranean health journal = La revue de santé de la Méditerranée orientale = al-Majallah al-sibhiyyah li-sharq al-mutawassat, 2016;22:327–34.
3. Arnott WP, Khadeejeh H, Moosmüller H. Towards aerosol light-absorption measurements with a 7-wavelength Aethalometer: evaluation with a photoacoustic instrument and 3-wavelength Nephelometer. Aerosol Sci Technol. 2005;39(1):17–29.
4. Ashbaugh LLM, Sadeh WC, Willy Z. A residence time probability analysis of sulfur concentrations at grand canyon national park. Atmos Environ (1967). 1985;19(8):1263–70.
5. Beegum SN, Moomth K, Babu SS, Satheesh SK, Vinoj V, Badarinath KVS, et al. Spatial distribution of aerosol black carbon over India during pre-monsoon season. Atmos Environ. 2009;43(5):1071–8.
6. Chen L, Zhang F, Yan P, Wang X, Sun L, Li Y, et al. The large proportion of black carbon (BC)-containing aerosols in the urban atmosphere. Environ Pollut. 2020;263:114507.
7. Cheng, D. (2018). Characteristics of black carbon aerosol and its influencing factors in typical cites of the PRD.
8. Dai ZL, Xiaoli, Wu P. Transport pathways and potential source region contributions of PM2.5 in Weifang: seasonal variations. Appl Sci. 2020;10:2835.
9. Database, N. E. S. (2016). Ministry of Environmental Protection (MEP) of China, Beijing.
10. Ding H. Effect of solar activity on earth’s climate and weather. Meteorol Monthly. 2019;45(3):297–304.
11. Drinovec L, Moćnik G, Zotter P, Prevot A, Ruckstuhl C, Coz E, et al. The “dual-spot” Aethalometer: An improved measurement of aerosol black carbon with real-time loading compensation. Atmos Meas Tech. 2015;8:1965–79.
12. Dumont M, Brun E, Picard G, Michou M, Lobois Q, Petit JR, et al. Contribution of light-absorbing impurities in snow to Greenland’s darkening since 2009. Nat Geosci. 2014;7(7):509–12.
13. Fan XC, Hongbin. Progress in observation studies of atmospheric aerosol radiative properties in China. Chin J Atmos Sci. 2013;37:477–98.
14. Guan D, Su X, Zhang Q, Peters GP, Liu Z, Lei Y, et al. The socioeconomic drivers of China’s primary PM 2.5 emissions. Environ Res Lett. 2014;9(2):024010.
15. Hansen ADAR, Novakov H, T. The aethalometer — An instrument for the real-time measurement of optical absorption by aerosol particles. Sci Total Environ. 1984;36:191–6.
16. Harrison RM, Beddows DCS, Jones AM, Calvo A, Alves C, Pio C. An evaluation of some issues regarding the use of aethelometers to measure woodsmoke concentrations. Atmos Environ. 2013;80:540–8.
17. Hua YT, Lili, Liu D. Source apportionment of black carbon aerosol during straw-burning period in spring and summer in Nanjing. Environ Sci Technol. 2017;1:147–55.
18. Hyvärinen AP, Vakkari V, Laakso L, Hooda R, Sharma VP, Panwar T, et al. Correction for a measurement artifact of the multi-angle absorption photometer (MAAP) at high black carbon mass concentration levels. Atmos Meas Tech. 2013;6:81–90.
19. Jing. Continuous observation of PM 2.5 and black carbon aerosol during summer in Beijing suburb. J Meteorol Sci. 2011;31:510–5.
20. Jing, Zhu B, Wang H, Yu X, An J, Kang H. Source apportionment of black carbon in different seasons in the northern suburb of Nanjing, China. Atmos Environ. 2019;201:190–200.
21. Liu Y, Yan C, Zheng M. Source apportionment of black carbon during winter in Beijing. Sci Total Environ. 2018;618:531–41.
22. Painter TJB, Ann C. Radiative forcing by light absorbing impurities in snow from MODIS surface reflectance data. Geophys Res Lett. 2012;39(17).
23. Peng X, Liu M, Zhang Y, Meng Z, Achal V, Zhou T, et al. The characteristics and local-regional contributions of atmospheric black carbon over urban and suburban locations in Shanghai, China. Environ Pollut. 2019;255:113188.
24. Pu W, Wang X, Wei H, Zhou Y, Shi J, Hu Z, et al. (2016). Properties of black carbon and other insoluble light-absorbing particles in seasonal snow of northwest China. Cryosphere Disc 1–59.
25. Ramanathan V, Carmichael G. Global and regional climate changes due to black carbon. Nat Geosci. 2008;1(4):221–7.
26. Ran L, Deng ZZ, Wang PC, Xia XA. Black carbon and wavelength-dependent aerosol absorption in the North China plain based on two-year aethalometer measurements. Atmos Environ. 2016;142:132–44.
27. Ravi Kiran V, Venkat Ratnam M, Krishna Murthy BV, Kant Y, Prasad P, Roja Raman M, et al. An empirical method for source apportionment of black carbon aerosol: results from Aethalometer observations at five different locations in India. Environ Pollut. 2019;254:112932.
28. Sandradewi J, Prevot A, Szidat S, Perron N, Alfarra M, Lanz V, et al. Using aerosol light absorption measurements for the quantitative determination of wood burning and traffic emission contributions to particulate matter. Environ Sci Technol. 2008;42:3316–23.
29. Seibert P, Kromp-Kolb H, Baltensperger U, Jost DT, Schwikowski M. Trajectory analysis of high-Alpine air pollution data. In: Gryning SE, Millán MM, editors. Air pollution modeling and its application X. Boston: Springer US; 1994. p. 595–6.
30. Streets DG, Gupta S, Waldhoff ST, Wang MQ, Bond TC, Yiyun B. Black carbon emissions in China. Atmos Environ. 2001;35(25):4281–96.
31. Sun T, He L, Zhang Y, Zeng L, Hu M, Huang X. Characteristics of the size distribution and mixing state of black carbon aerosol in Shenzhen in winter. Chin Sci Bull. 2011;56(21):1703–10.
32. Tianle S, Lingyan H, Long H, Yanling L, Xin Z, Mingdi Z, et al. The vertical distribution of atmosphere pollutants in Shenzhen in winter. Acta Sci Circumst. 2019;39(1):64–71.
33. Wang X, Pu W, Ren Y, Xuelei Z, Zhang X, Shi J, et al. Observations and model simulations of snow albedo reduction in seasonal snow due to insoluble light-absorbing particles during 2014 Chinese survey. Atmos Chem Phys. 2017;17:2279–96.
34. Wu D, Wu C, Liao B, Chen H, Wu M, Li F, et al. Black carbon over the South China Sea and in various continental locations in South China. Atmos Chem Phys. 2013;13:12257–70.
35. Wu. Observation of black carbon aerosol in Beijing during the summer of 2005. J Atmos Environ Opt. 2006;1:210–5.
36. Wu. Quantifying black carbon light absorption enhancement by a novel statistical approach. Atmos Chem Phys. 2018;18:289–309.
37. Yang Y, Ni C, Jiang M, Chen Q. Effects of aerosols on the atmospheric boundary layer temperature inversion over the Sichuan Basin, China. Atmos Environ. 2021;262:118647.
38. Zhang Y, Li J, Zhang G, Zotter P, Huang R-J, Tang J, et al. Radiocarbon-based source apportionment of carbonaceous aerosols at a regional background site on Hainan Island, South China. Environ Sci Technol. 2014;48:2651–9.
39. Zhi G, Chen Y, Feng Y, Xiong S, Li J, Zhang G, et al. Emission characteristics of carbonaceous particles from various residential coal-stoves in China. Environ Sci Technol. 2008;42:3310–5.
40. Zhuang BL, Wang TJ, Liu J, Li S, Xie M, Yang XQ, et al. Continuous measurement of black carbon aerosol in urban Nanjing of Yangtze River Delta, China. Atmos Environ. 2014;89:415–24.
41. Zotter P, Herich H, Gysel M, El-Haddad I, Zhang Y, Močnik G, et al. Evaluation of the absorption Ångström exponents for traffic and wood burning in the Aethalometer-based source apportionment using radiocarbon measurements of ambient aerosol. Atmos Chem Phys. 2017;17:4229–49.

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