Since January 2020 Elsevier has created a COVID-19 resource centre with free information in English and Mandarin on the novel coronavirus COVID-19. The COVID-19 resource centre is hosted on Elsevier Connect, the company's public news and information website.

Elsevier hereby grants permission to make all its COVID-19-related research that is available on the COVID-19 resource centre - including this research content - immediately available in PubMed Central and other publicly funded repositories, such as the WHO COVID database with rights for unrestricted research re-use and analyses in any form or by any means with acknowledgement of the original source. These permissions are granted for free by Elsevier for as long as the COVID-19 resource centre remains active.
Substantial decreases of light absorption, concentrations and relative contributions of fossil fuel to light-absorbing carbonaceous aerosols attributed to the COVID-19 lockdown in east China

Yu-Chi Lin, Yan-Lin Zhang, Feng Xie, Mei-Yi Fan, Xiaoyan Liu

A Yale-NUIST Center on Atmospheric Environment, International Joint Laboratory on Climate and Environment Change, Nanjing University of Information Science and Technology, Nanjing, 210044, China

b Key Laboratory Meteorological Disaster, Ministry of Education & Collaborative Innovation Center on Forecast and Evaluation of Meteorological Disaster, Nanjing University of Information Science and Technology, Nanjing, 210044, China
c Jiangsu Provincial Key Laboratory of Agricultural Meteorology, School of Applied Meteorology, Nanjing University of Information Science & Technology, Nanjing, 210044, China

**Article info**

**Abstract**

To prevent the spread of Coronavirus disease-2019 (COVID-19), China adopted the lockdown measures in late January 2020, providing a platform to study the response of air quality and atmospheric chemical and physical properties to strict reduced emissions. In this study, the continuous measurements of aerosol light absorption were conducted in Nanjing, east China, from January 3 to March 31, 2020. Our results showed that the contribution of black carbon (BC) to light absorption at the different wavelengths was more than 75% and the rest light absorption was contributed by brown carbon (BrC), which was mainly originated from primary emissions. Secondary BrC absorption, which was mainly produced by photochemical oxidation, constituted a minor fraction (2-7%) of the total absorption. Compared with the sampling in the pre-lockdown, the significant decreases of BC (43%) and secondary BrC absorption (31%) were found during the lockdown period, resulting in a substantial decrease of solar energy absorbance by 36% on a local scale. The control measures also changed the diurnal variations of light absorption. Due to the reduced emissions, the relative fraction of fossil fuel to BC also dropped from 78% in the pre-lockdown to 71% in the lockdown. The concentrations of BC, PM2.5 and NO2 decreased 1.1 μg m⁻³, 33 μg m⁻³ and 9.1 ppb whereas O3 concentration increased 9.0 ppb during the COVID-19 lockdown period. The decreased concentrations of BC, PM2.5 and NO2 were mainly contributed by both emission reduction (51-64%) and meteorological conditions (36-49%). Our results highlighted that the balance of control measures in alleviation of particulate matter (PM) and O3 pollution, and meteorology should be seriously considered for improvement of air quality in this urban city of China.

© 2021 Elsevier Ltd. All rights reserved.

**1. Introduction**

The first case of novel coronavirus disease-2019 (COVID-19) was found in Wuhan, China in 2019 December (Guan et al., 2020; Wu et al., 2020). The COVID-19 was caused by severe acute respiratory syndrome coronavirus 2 (SARS-2) (Wu et al., 2020). It was not only found in China, but also spread to other countries, resulting in global pandemic due to its human-to-human transmissibility (Hui et al., 2020; Wang et al., 2020a). Until now, more than 96 million cases of COVID-19 have been reported and caused more than 2,098,000 deaths (https://covid19.who.int). To prevent fast spread of COVID-19, China imposed nationwide restriction of movement in its population in late January 2020 (Tian et al., 2020). The strict lockdown meant shutdown of all industrial activities and travels. Schools and entertainment venues have also been closed. Due to the reduced emissions, the relative fraction of fossil fuel to BC also dropped from 78% in the pre-lockdown to 71% in the lockdown. The concentrations of BC, PM2.5 and NO2 decreased 1.1 μg m⁻³, 33 μg m⁻³ and 9.1 ppb whereas O3 concentration increased 9.0 ppb during the COVID-19 lockdown period. The decreased concentrations of BC, PM2.5 and NO2 were mainly contributed by both emission reduction (51-64%) and meteorological conditions (36-49%). Our results highlighted that the balance of control measures in alleviation of particulate matter (PM) and O3 pollution, and meteorology should be seriously considered for improvement of air quality in this urban city of China.
and SO2) due to the lockdown in northern China varied from 16 to 54%. The declined concentrations of these primary pollutants were mainly attributed to the reduced emissions from traffic and industrial sources. Nevertheless, the reduction of NOx emissions enhanced O3 concentrations. In the Yangtze River Delta (YRD) region, the significant reduced emissions from industries (reduction rate from 20% for NOx to 50% for VOCs), vehicle traffic (~75% for primary air pollutants) and construction (~90% for particulate matters) during the lockdown period decreased PM2.5, NO2 and SO2 by 32, 45 and 20%, respectively (Li et al., 2020).

Aerosol light absorption influences significantly on atmospheric visibility and Earth’s climate change (Watson, 2002; Ramanathan and Carmichael, 2008). Black carbon (BC) and brown carbon (BrC) are important light-absorbing species in atmospheric aerosols. Both species are also the largest sources of uncertainty in predicting future climate due to their limited understanding of emissions, formation, light properties and their interactions (Bond et al., 2013). BC absorbs solar radiation over a broad spectrum range, from ultraviolet (UV) to infrared (IR) with absorption Ångström exponent (AAE) of close to unity (Bond et al., 2013). In contrast, BrC has strong light absorption from near-UV to short IR with AAE of ~1; its light absorption usually decreases from near-UV to IR (Lack and Langridge, 2013). Light absorption by BC and BrC changes the radiative forcing, resulting in warming effects on Earth system. On average, the global radiative forcing caused by BrC was 0.22–0.57 (W m$^{-2}$), which was 25–75% of that by BC (Lin et al., 2014).

Previous numerous studies have shown that combustion of fossil fuel and biomass burning (BB) were major sources of BC aerosols (Zhao et al., 2012; Cheng et al., 2013; Zhang et al., 2017; Ni et al., 2018). Although the origins of BrC are not well characterized, biomass burning, coal combustion and vehicle emissions along with biogenic release of fungi, plant debris and humic matter are considered important sources of primary BrC (Andreae and Crutzen, 1997; Rizo et al., 2011, 2013; Yan et al., 2017; Zhang et al., 2017; Yuan et al., 2019). Secondary BrC can be produced by multi-phase reactions between gas-phase, particles and in-cloud processes (Harrison et al., 2005; Nozière et al., 2007; De HANN et al., 2009). Due to complete shutdown of economic activities, the COVID-19 lockdown provided us an platform to study the response of air quality, chemical and physical properties of aerosols to strict reduced anthropogenic emissions over China. In this work, the continuous measurements of aerosol light absorption of PM2.5 were conducted in Nanjing city from January 3 to March 31, 2020. During this period, Nanjing city implemented “lockdown” policies, which indicates that people have to staying at home and all economic activities are shut down, from February 3 to 19, in order to prevent spreads of COVID-19. After February 20, 80% of the economic activities, such as public transportation and industries, re-operated. Here, we compared the aerosol light absorption before (January 3 to February 2), during (February 3 to 19) and after (February 20 to March 31) the COVID-19 lockdown. The comparisons gave us an insight for understanding the influence of strict reduced anthropogenic emissions on aerosol light absorption in this urban city. Using the observed data, the solar energy absorbance by light-absorbing carbonaceous aerosols were also calculated. Our result provided a hint to understand the impacts of COVID-19 lockdown on the optical properties, concentrations, sources and solar energy absorbance by BC and BrC in a polluted city over the YRD region of China. On the other hand, the lockdown supplied a platform to explore the response of air pollution to reduced anthropogenic emissions. Thus, our results provided valuable information for policy-maker to formulate the control measures in improvement of air quality.

2. Method

2.1. Sampling site

Continuous aerosol light absorption was monitored using an Aethalometer in Nanjing, China, from January 3 to March 31, 2020. During the sampling period, the Aethalometer was installed inside the monitoring station, which is located at the rooftop of a seven-story building on the campus of Nanjing University of Information Science and Technology (NUIST, 32.21°N, 118.72°E, shown in Fig. 1). The sampling site is encompassed by local roads and an expressway. In addition, petroleum chemical refineries and steel manufacturing plants are situated in the northeast and east direction with a distance of approximately 5 km. Thus, vehicle and industrial emissions were the dominant sources of air pollution at the sampling site. During the sampling period, the hourly data of PM2.5 mass, CO, NO2, SO2 and O3 along with ambient temperature (T) and relative humidity (RH) were acquired from the Pukou air quality monitoring station which is located to the southwest of the receptor site.

2.2. Principle of Aethalometer measurement

In this work, an Aethalometer (model AE33, Magee Scientific) was used to monitor aerosol light absorption ($b_{\text{abs}}(\lambda)$). This instrument has been widely used to continuously measure aerosol light absorption at seven wavelengths (λ), including 370, 470, 520, 590, 660, 880 and 950 nm (Drinovec et al., 2015; HELIN et al., 2018; JING et al., 2019; WANG et al., 2019). During the sampling period, the flow rate of AE33 was set to 5 L min$^{-1}$ and the inlet (model SCC1.829, BGI) cut-off size was 2.5 μm. Briefly, aerosol particles were continuously collected on the filter tapes and the optical attenuation (ATN) was measured with a time resolution of 1 min. Subsequently, the aerosol light absorption can be calculated by ATN values, also, BC concentrations can be estimated by ATN and the mass absorption cross section (MAC = 7.8 m$^{2}$ g$^{-1}$) at 880 nm since the light absorption at this wavelength associated with other particle species, such as BrC, is insignificant (Drinovec et al., 2015). The principle of the estimation of light absorption by the Aethalometer can be seen in supplement Text S1 and more details of AE33 can be found elsewhere (Drinovec et al., 2015). In addition to carbonaceous aerosols, dust is also an important light-absorbing species in the atmosphere. A previous study have suggested that the contribution of dust particles to light absorption was less than 5% in the supermicron PM at an urban site over the Northern China Plain (NCP) region (YANG et al., 2009). Nevertheless, the proportion of light absorption by dust in aerosols with the dynamic diameter of larger than 1.0 μm was approximately 10% of that by BC aerosols (CLARK et al., 2004). This did indicate that using the Aethalometer for monitoring the light absorption by carbonaceous aerosols, such as BC and BrC, exhibited a positive artifact, which was caused by dust particles.

2.3. Estimation of light absorption due to primary and secondary BrC

In this section, we attempted to partition BC and BrC light absorption from our observed data. Due to existence of dust particles, BC and BrC light absorption might be overestimated by Aethalometer measurements. However, we did not have other measurements to correct the positive artifact. Consequently, the influence of dust particles on BC and BrC light absorption was not considered in the estimation. Based on the assumption, the secondary BrC light absorption ($b_{\text{abs,BrC-secondary}}(\lambda)$) can be calculated as (WANG et al., 2019):
where \((b_{\text{abs}}(\lambda)/[BC])_{\text{pri}}\) is the \(b_{\text{abs}}(\lambda)\)-to-BC ratio in primary emission sources at wavelengths of 370, 470, 520, 590 and 660 nm with a unit of \(\text{m}^2\ \text{g}^{-1}\). \([BC]\) is the concentration of particulate BC (\(\mu\text{g m}^{-3}\)), which is retrieved from the relationship between the ATN and MAC at a wavelength of 880 nm. The ratio of \((b_{\text{abs}}(\lambda)/[BC])_{\text{pri}}\) varied with the different emission sources and therefore we used a minimum R-squared (MRS) approach to obtain the appropriate values of \((b_{\text{abs}}(\lambda)/[BC])_{\text{pri}}\) (Wang et al., 2019). First, the arbitrary values of \((b_{\text{abs}}(\lambda)/[BC])_{\text{pri}}\) from 0 to 100 with an increasing increment of 0.1 at the individual wavelengths were tested to obtain the \(b_{\text{abs,BrC-sec}}(\lambda)\) values. Further, a coefficient of determination \((R^2)\) was calculated by linear regression between \(b_{\text{abs,BrC-sec}}(\lambda)\) and BC concentration at the individual wavelengths. The appropriate \((b_{\text{abs}}(\lambda)/[BC])_{\text{pri}}\) value at each wavelength was then acquired when the minimum \(R^2\) value between \(b_{\text{abs,BrC-sec}}(\lambda)\) and BC was found (see in Figure S1). After acquiring the \((b_{\text{abs}}(\lambda)/[BC])_{\text{pri}}\) values, we can then calculate the \(b_{\text{abs,BrC-sec}}(\lambda)\) values at the different wavelengths of each sampling. Details of this approach can be found elsewhere (Wang et al., 2019).

After obtaining the \(b_{\text{abs,BrC-sec}}(\lambda)\), the light absorption of primary BrC \((b_{\text{abs,BrC-pri}}(\lambda))\) can then be calculated as (Wang et al., 2019):

\[
b_{\text{abs,BrC-pri}}(\lambda) = b_{\text{abs,BrC}}(\lambda) - b_{\text{abs,BrC-sec}}(\lambda)
\]

where \(b_{\text{abs,BrC}}(\lambda)\) is the light absorption by total BrC aerosols. Details of the estimation of \(b_{\text{abs,BrC}}(\lambda)\) can be found in *Supplement Text S2*.

3. Results and discussion

3.1. Light absorption of BC and BrC

During the sampling period, the average temperature at the receptor site was 9.1 ± 6°C with higher values after mid-March (Fig. 2). The relative humidity averaged at 70 ± 23% with significant fluctuations on an hourly basis. Northerly and southerly wind prevailed at the sampling site with an hourly average wind speed of 1.7 ± 0.9 m s\(^{-1}\). Precipitation occurred occasionally while heavy rainfall was observed on March 26 with 6-h accumulated rainfall of higher than 25 mm. Fig. 3 plots the relationship between the average aerosol light absorption at the different wavelengths in Nanjing during the different periods. Here, a power law function \((b_{\text{abs}}(\lambda) = \lambda^{-\alpha_{\text{BC}}}}\) was used to fit the data between light absorption and wavelength. The results revealed that the AAE values varied from 1.3 to 1.4, indicating the presence of both BC and BrC aerosols in the atmosphere. Note that the light absorption at the different wavelengths during the lockdown was approximately two-third of that in the pre-lockdown. After lockdown, the increases of light absorption were found, but the values were still lower than those in the pre-lockdown. The significant decrease of light absorption during the lockdown might be attributed to both reduced anthropogenic emissions and meteorological conditions and this will be discussed later.

Figure S2 shows the time series of aerosol light absorption by BC, primary and secondary BrC at the different wavelengths. In this figure, the partitioned BC and BrC absorption from the total light absorption was estimated by Eqs. (S6) and (S7); the assumed AAEC value would cause the uncertainty of estimated BC and BrC light absorption. Previously, Kirchstetter et al. (2004) measured the aerosol light absorption near a traffic road and in a tunnel, and they suggested that the AAEC value was in a range of 0.6–1.3. In this work, the AAEC was assumed to be 1.0 for the estimation (Laskin et al., 2015). This resulted in the uncertainty of estimated BC light absorption less than 29% at the different wavelengths compared with those with AAEC of 0.6 or 1.3; the uncertainty decreased with increasing wavelengths (see in Table S1). Since BrC light absorption is obtained by the difference between the total and partitioned BC light absorption, the uncertainty of BrC light absorption should be the same as that of BC. Moreover, we used MRS method to estimate the secondary BrC light absorption and the uncertainty of MRS approach for isolation of one parameter (e.g. secondary BrC light absorption) from another parameter (e.g. total light absorption) was less than 23% (Wu and Yu, 2016).

On average, the total aerosol light absorption \((b_{\text{abs, total}})\) was 52 ± 31, 38 ± 23, 33 ± 20, 28 ± 17, 23 ± 14, 17 ± 10 and 15 ± 9 Mm\(^{-1}\) at wavelengths of 370, 470, 520, 590, 660, 880 and 950 nm, respectively (see in Table 1). Obviously, BC was a dominant contributor to light absorption with a relative fraction of higher
than 75%. In terms of BrC, the average light absorption was 13 ± 5, 7.0 ± 3, 5.0 ± 2, 3.0 ± 1 and 1.0 ± 0.7 Mm$^{-1}$ at wavelengths of 370, 470, 520, 590 and 660 nm, which accounted for 25%–5% of the total light absorption at these wavelengths. The aerosol light absorption of primary BrC exceeded that of secondary BrC by factors of 1.5–2.5 at the different wavelengths, suggesting that primary emission was likely the main source to BrC absorption in Nanjing. The MAC of BrC was very distinctive due to its various emission sources (e.g. the MAC of BrC was from 0.21 m$^2$ g$^{-1}$ for wood burning to 2.3 m$^2$ g$^{-1}$ for kerosene at $\lambda$ of 370 nm, Olson et al., 2015) and therefore we cannot estimate the absolute concentrations of BrC by using ATN and MAC. The absorption of BrC at 370 nm was the most intensive than those at other wavelengths. Consequently, the BrC absorption at $\lambda = 370$ nm was considered the representative value of BrC absorption (Wang et al., 2019) and was subsequently used to explore the optical properties of BrC in this study.

The light absorption at $\lambda = 880$ nm can be considered as the representative value of BC absorption and therefore we used the light absorption at $\lambda = 880$ nm to discuss the variations of BC
absorption (Yang et al., 2009; Drinovec et al., 2015). As listed in Table 2, the average BC absorption in the pre-lockdown was 21 ± 12 Mm⁻¹/C₀, which was 1.8 times higher than that (12 ± 6 Mm⁻¹/C₀) during the lockdown period. Using the one-way ANOVA test, a significant difference (p < 0.05, 95% confidence interval) of BC absorption was found between the pre- and during-lockdown. The BC absorption decreased by 43% during the lockdown period was the response to the declined BC concentrations by the strict reduction of anthropogenic emissions and this will be discussed in the following section. After the lockdown, the economic activities re-operated, resulting in an increase of BC absorption. The light absorption by primary BrC in the pre-lockdown was 8.6 ± 9 Mm⁻¹, which was insignificant different from those in the pre- (9.4 ± 6 Mm⁻¹, p > 0.05) and after-lockdown (8.7 ± 9 Mm⁻¹, p > 0.05). In terms of secondary BrC, its absorption during the lockdown period was 2.4 ± 3 Mm⁻¹, which was significant (p < 0.05) lower than those in the pre- (3.5 ± 4 Mm⁻¹) and after-lockdown (3.0 ± 4 Mm⁻¹). In conclusion, the COVID-19 lockdown did changes the aerosol light absorption by BC and secondary BrC. On average, the light absorption of BC and secondary BrC due to lockdown in Nanjing decreased by 43 and 31%, respectively.

![Fig. 3. Aerosol light absorption at the different wavelengths in Nanjing during the different sampling periods. The circles and solid lines denote mean values and standard deviations. The average AAE values with uncertainty are also shown.](image)

**Table 1**

Average aerosol light absorption of total aerosol (b_{abs, total}), BC (b_{abs,BC}), primary (b_{abs,BrC-pri}) and secondary BrC (b_{abs,BrC-sec}) at the different wavelengths in Nanjing during the sampling period.

| Parametersa | Wavelength (nm) | 370 | 470 | 520 | 590 | 660 | 880 | 950 |
|-------------|----------------|-----|-----|-----|-----|-----|-----|-----|
| b_{abs, total} | 52 ± 31 | 38 ± 23 | 33 ± 20 | 28 ± 17 | 23 ± 14 | 17 ± 10 | 15 ± 9 |
| b_{abs,BC} | 39 ± 24 (75%) | 31 ± 19 (82%) | 28 ± 17 (85%) | 25 ± 15 (89%) | 22 ± 13 (95%) | 17 ± 10 (100%) | 15 ± 9 (100%) |
| b_{abs,BrC-pri} | 9.3 ± 8 (18%) | 5.0 ± 4 (13%) | 3.3 ± 2 (10%) | 2.0 ± 2 (7%) | 0.6 ± 1 (3%) | 0 | 0 |
| b_{abs,BrC-sec} | 3.7 ± 4 (7%) | 2.0 ± 2 (5%) | 1.7 ± 1 (5%) | 1.0 ± 0.8 (4%) | 0.4 ± 0.5 (2%) | 0 | 0 |

a The unit of aerosol light absorption is Mm⁻¹.

b The data on the parentheses denote the relative fractions of BC, primary BrC and secondary BrC to the total aerosol light absorption.
3.2. Diurnal cycles and formation pathways of secondary BrC

The diurnal cycles of light absorption of BC, primary and secondary BrC are plotted in Fig. 4. The diurnal cycles of light absorption of BC and BrC can be influenced by emission sources, dynamics of boundary layer height (BLH) and formation mechanisms (secondary BrC only). In the pre-lockdown, BC absorption showed relative flat values (20–21 Mm$^{-1}$) from nighttime to early morning hours and then increased from 6:00 local time (LT) to 9:00 LT. After peaking at 9:00 LT (24 Mm$^{-1}$), BC absorption decreased gradually and reached a minimum value at 16:00 LT (~18 Mm$^{-1}$), then increased and kept higher absorption in the evening (20–21 Mm$^{-1}$). The peak of BC absorption during the morning traffic rush hours was associated with intensive vehicle emissions since peaks of CO and NO$_2$ were also found (Fig. S3). The lower BC absorption from midday to the afternoon might be associated with the high BLH, which was favorable for dilution of air pollutants. In this work, we calculated the height at the dry adiabatic process converted to wet adiabatic process (seen in Supplement Text S3). This can be considered the tendency of the BLH to some extent (namely proxy of boundary layer height, PBLH, afterwards). As shown in Figure S3, the higher PBLH was regularly found from midday to the afternoon, increasing the atmospheric dilution capability for air pollutants and lowering the concentrations and light absorption of BC aerosols.

During the lockdown period, the BC absorption decreased and the peak on the traffic rush hours was not observed. After the lockdown, the BC absorption increased and the peak on the traffic rush hours was found again. These findings implied that strict control of vehicle emissions not only decreased the BC absorption, but also changed its diurnal cycles. For primary BrC, its absorption (8.6 Mm$^{-1}$) during the lockdown period was slightly lower than that (9.4 Mm$^{-1}$) in the pre-lockdown and its peak on the traffic rush hours disappeared. This also indicated that reduced vehicle emissions changed the diurnal cycles of primary BrC absorption. In terms of secondary BrC absorption ($b_{abs,BrC-sec}$ in Fig. 4), the significant diurnal variations were found with higher values during the midday to evening. The diurnal cycles of $b_{abs,BrC-sec}$ can be explained by dilution dynamic of BLH and potential formation processes. Here, we used a function of $b_{abs,BrC-sec} \times $ PBLH to eliminate the effect of dilution factor on secondary BrC absorption and attempted to explore the potential formation pathway of secondary BrC. Odd oxygen ($O_x = NO_2 + O_3$) can be considered an index of photochemical oxidation capability. As shown in Fig. 4, the peaks of Ox concentrations and temperature were observed around 16:00 LT, suggesting strong photochemical oxidation. In general, the maximum of $b_{abs,BrC-sec} \times $ PBLH were usually found 2 h (~18:00 LT) later than the peaks of Ox and temperature. Meanwhile, a positive correlation ($R = 0.79$ to 0.89, $p < 0.05$) was found between $b_{abs,BrC-sec} \times $ PBLH and Ox, but a negative correlation ($R = -0.72$ to $-0.89$) was found between $b_{abs,BrC-sec} \times $ PBLH and RH (Fig. 5). This eluci- dated that secondary BrC in Nanjing was predominantly from photochemical reactions rather than aqueous-phase processes. Our result was in agreement with those in Xianghe over the NCP region and Hong Kong (Wang et al., 2019; Zhang et al., 2020) where the production of BrC was mainly from photochemical oxidation. On the other hand, the significant increase of $b_{abs,BrC-sec} \times $ PBLH was found in the after-lockdown, suggesting enhanced secondary BrC absorption, which might be attributed to the stronger photochemical reactions (high Ox concentrations) due to the higher atmospheric temperature conditions.

3.3. Concentrations and sources of BC

Assuming the MAC was 7.8 m$^2$ g$^{-1}$, the BC concentrations can be estimated by using the BC light absorption at $\lambda$ of 880 nm (Drinovec et al., 2015). During the entire sampling period, the hourly concentration of BC varied from 0.1 to 8.0 mg m$^{-3}$, with a mean value of 2.2 ± 0.8 mg m$^{-3}$ (Fig. 2). Significant differences of BC concentrations were found between, during and after the lockdown periods. On average, the BC concentration was 1.6 ± 0.8 mg m$^{-3}$ during the lockdown period, which was much lower than those in the pre- (2.7 ± 2.0 µg m$^{-3}$) and after-lockdown (2.1 ± 1.0 µg m$^{-3}$) (see in Table 2). The decrease of BC concentration during the lockdown was ascribed to emission reduction since all economic activities were closed. Apart from emission sources, BC concentrations are also influenced by meteorological conditions, such as BLH, which can reflect the atmospheric dynamic of dilution for air pollutants. On average, the PBLH was 0.44, 0.51 and 0.96 km in the pre-, during- and after-lockdown (see in Table 2). Compared to the PBLH in the pre-lockdown, a higher value was found in the lockdown, suggesting that the decrease of BC concentration in the lockdown was also attributed to higher boundary layer in addition to reduced emissions.

Fossil fuel and biomass burning are important sources of atmospheric particulate BC in China. Using Aethalometer model, the relative contribution of fossil fuel and BB to airborne BC can be quantified (Sangradaswi et al., 2008; Helin et al., 2018; Diaz Resquin et al., 2018; Dumka et al., 2018). The details of Aethalometer model can be found in supplement Text S4. Fig. 6 shows the time series of absolute BC concentrations contributed by fossil fuel and BB emission sources. The relative contributions of fossil fuel and BB to BC are also plotted in this figure. During the entire sampling period, 77% of BC was contributed by fossil fuel emissions while 23% was from biomass burning, indicating that fossil fuel was a dominant emission source of BC in Nanjing in the winter and spring seasons. The contribution of fossil fuel to BC decreased from 78% in the pre-lockdown to 71% during the lockdown period. After the lockdown, the relative contribution of fossil fuel rose up to 78%. In Aethalometer model, the AAEE values of fossil fuel (AAE$_{FF}$) and biomass burning (AAE$_{BB}$) affect the partitions of the different sources to BC.

Table 2

| Species/parameters | Before | Lockdown | After |
|--------------------|--------|----------|-------|
| BC absorption (Mm$^{-1}$) | 21 ± 12 | 12 ± 6 | 16 ± 9 |
| Primary BrC absorption (Mm$^{-1}$) | 9.4 ± 6 | 8.6 ± 9 | 8.7 ± 2 |
| Secondary BrC absorption (Mm$^{-1}$) | 3.5 ± 4 | 2.4 ± 3 | 3.0 ± 4 |
| PM$_{2.5}$ (µg m$^{-3}$) | 58 ± 39 | 25 ± 15 | 31 ± 19 |
| BC (µg m$^{-3}$) | 2.7 ± 2 | 1.6 ± 0.8 | 2.1 ± 1 |
| CO (ppm) | 0.58 ± 0.4 | 0.43 ± 0.3 | 0.5 ± 0.3 |
| NO$_2$ (ppb) | 18 ± 11 | 8 ± 6 | 17 ± 10 |
| SO$_2$ (ppb) | 17 ± 3 | 1.8 ± 0.6 | 21 ± 1 |
| O$_3$ (ppb) | 18 ± 13 | 27 ± 11 | 29 ± 17 |
| T (°C) | 4.9 ± 4 | 7.3 ± 6 | 13 ± 6 |
| RH (%) | 80 ± 19 | 72 ± 22 | 61 ± 24 |
| WS (m s$^{-1}$) | 1.7 ± 0.8 | 1.6 ± 1 | 1.8 ± 0.9 |
| PBLH (km) | 0.44 ± 0.5 | 0.51 ± 0.5 | 0.96 ± 0.7 |
| E$_{BC}$ (W m$^{-2}$) | 22 ± 12 | 13 ± 7 | 13 ± 5 |
| E$_{BC-sec}$ (W m$^{-2}$) | 2.3 ± 1 | 2.0 ± 2 | 2.0 ± 2 |
| E$_{sec}$ (W m$^{-2}$) | 0.9 ± 0.9 | 0.7 ± 0.8 | 0.7 ± 0.9 |
| Ext$_{sec}$ (W m$^{-2}$) | 25 ± 13 | 16 ± 8 | 16 ± 9 |
significant differences of the results between the various selected AAE pairs. However, in this work, the changes of relative contributions of fossil fuel to BC in the pre-, during- and post-lockdown events were much more concerned rather than evaluating their absolute values. Thus, we selected specific AAE values (AAE$_{BB}$ = 2.0 and AAE$_{FF}$ = 1.0) in the partitions of emission sources to BC. Although uncertainty was existed in the estimation, our results did highlight that the shutdown of vehicle and industries significantly decreased the relative contribution of fossil fuel to BC aerosols.

3.4. Influence of reduced emission on BC concentrations

During the COVID-19 lockdown period, significant decreases of BC concentrations were observed. Meanwhile, the higher temperature, lower RH and high PBLH were also found (Table 2). This suggested that the declined BC concentrations were probably attributed to changes of meteorological conditions and reduced emissions. In this section, we tried to estimate the relative contributions of meteorological conditions and reduced emissions to the changes of BC concentrations during the lockdown period. Boundary layer height is a direct factor controlling the ambient levels of primary air pollutants and therefore BLH would be considered in the estimation. Assuming the changes in concentrations of BC in the lockdown were associated with the reduced emissions ($C_E$) and changes of meteorological conditions ($C_M$), the total changing concentrations of BC ($C_T$) can be simply expressed as:

$$C_T = C_M + C_E$$  \hspace{1cm} (3)
metereological parameter influencing the variations of concentrations in air pollutants. As listed in Table 2, the BC concentrations in the pre- and during-lockdown were 2.7 and 1.6 $\mu$g m$^{-3}$, respectively, indicating that BC concentrations decreased 1.1 $\mu$g m$^{-3}$ in the COVID-19 lockdown. The PBLH during the lockdown period was 0.51 km, which was approximately 1.2 times higher than that in the pre-lockdown (0.44 km). This demonstrated that BC concentrations during the lockdown period should decrease by 20% due to the dilution factor without any changes of emissions since the PBLH was higher. Thus, the decreased BC concentrations associated with meteorological conditions (without considering reduced emissions) during the lockdown period was then estimated to be 0.54 $\mu$g m$^{-3}$ ($C_2 = 0.20 \times 2.7 \mu$g m$^{-3}$), which accounted for 49% of the total decreased BC concentrations. In other words, the decreases of BC concentrations associated with emission control was 0.56 $\mu$g m$^{-3}$, contributing for 51% to the total decreased BC (see in Table 3). Using the same approach, we also estimated the decreased concentrations by reduced emissions and changes of meteorology for other air pollutants. Compared to those in the pre-lockdown, the concentrations of PM$_{2.5}$ decreased approximately 33 $\mu$g m$^{-3}$ during the lockdown period. Meanwhile, a significant decrease was also found for NO$_2$ (by 9.1 ppb, $p < 0.05$). On the contrary, O$_3$ increased about 9.0 ppb during the lockdown period. Using Eq. (3), we estimated that the decreased concentrations of PM$_{2.5}$ and NO$_2$ by reduced emissions were 21 $\mu$g m$^{-3}$ and 5.5 ppb, respectively. The reduced emission increased the O$_3$ concentrations by approximately 13 ppb. The enhancement of O$_3$ concentration might be attributed to the decreases of NOx concentration since high NOx concentration would inhibit the formation of atmospheric ozone (Ding et al., 2013). Besides, a decrease of PM$_{2.5}$ concentration might be another reason for enhancements of O$_3$ levels (Li et al., 2019). Considering the effect of atmospheric dilution dynamics, the relative contributions of emission control to the decreases of BC, PM$_{2.5}$ and NO$_2$ were 51, 64 and 60%, respectively. The relative contributions of meteorological conditions to the decreased concentrations of these air pollutants varied from 36 to 49%. Our results examined that meteorological condition played an important role in controlling the concentrations of air pollutants. Therefore, meteorology should be considered when the control measures for improvement of air quality were made, especially in alleviating haze formation when the atmospheric air conditions were usually stagnant with high relative humidity environment, which facilitate the formation of secondary aerosols (Du et al., 2020).

3.5. Influence of COVID-19 lockdown on solar energy absorbance

The changes of light absorption due to the COVID-19 lockdown would be expected to change direct solar energy absorbance in the atmosphere. Here, we used a simplistic model (Supplement Text S5) to estimate the solar energy absorbance by BC ($E_{BC}$) and BrC ($E_{BrC}$) at wavelengths from 300 to 950 nm before, during and after the COVID-19 lockdown (Bosch et al., 2014; Liu et al., 2019). Based on the aerosol light absorption, we estimated that the total solar energy absorbance during the entire sampling period varied from 1.2 to 67 W m$^{-2}$, with a mean value of 20 ± 12 W m$^{-2}$. The amount of solar energy absorbance by BrC relative to BC ($E_{BrC}/E_{BC}$) was 17 ± 0.7%. If we calculated the solar energy absorbance at wavelengths from 300 to 460 nm, we found that the total solar energy absorbance was 5.9 ± 3 W m$^{-2}$ and the average $E_{BrC}/E_{BC}$ ratio was 28 ± 13%. This illustrated that the warming effects in the global atmosphere by BrC was more important in short-wavelength range. Subsequently, the changing ratios of solar energy absorbance by BC and BrC can be estimate by the differences of $E_{BC}$ and $E_{BrC}$ before and during the lockdown periods. Before the COVID-19 lockdown, the solar energy by BC, primary and secondary BrC were 22 ± 12, 2.3 ± 1 and 0.9 ± 0.9 Wm$^{-2}$, respectively. The declined solar energy absorbance ($E_{BrC} = 13 + 7$ Wm$^{-2}$, $E_{BrC-pri} = 2.0 + 2$ Wm$^{-2}$ and $E_{BrC-sec} = 0.7 + 0.8$ Wm$^{-2}$) was found during the lockdown period (Table 2), resulting in the changing ratio of total solar energy absorbance (−36%) by light-absorbing carbonaceous aerosols in Nanjing (Figure S4). This highlighted that the strict reduction of anthropogenic emissions during the COVID-19 lockdown did decrease solar energy absorbance on a local scale.

4. Conclusions

In this study, we continuously monitored the aerosol light absorption at seven wavelengths ($\lambda$) in Nanjing, China, from January 3 to March 31, 2020 and the data was used to evaluate the impact of COVID-19 lockdown on the aerosol light absorption. Our results showed that BC was the major absorbing carbonaceous aerosols, accounting for 75–100% of the total light absorption at the different wavelengths. The rest of absorption was contributed by BrC, which was mainly originated from primary emissions. Compared to the pre-lockdown, the light absorption of BC and secondary BrC
decreased by 43% and 31%, respectively, causing a decrease of total solar energy absorbance by 36%. On the other hand, substantial decreases in concentrations of BC (1.1 μg m⁻³), PM₂.₅ (33 μg m⁻³) and NO₂ (9.1 ppb) were found during the lockdown period. On the contrary, O₃ concentration enhanced by 9.0 ppb in the COVID-19 lockdown. Consideration the dilution dynamic by boundary layer, we estimated that the strict reduced emissions decreased the concentrations of BC, PM₂.₅ and NO₂ by 51–64% while meteorology contributed 36–49% to the decreases.

The COVID-19 was announced an emergency by the World Health Organization (WHO) since its human-to-human transmissibility and global pandemic, threatening seriously human's life. To prevent spread of the virus, Chinese government was the first country to implement the “lockdown policy” in many cities. This provided us a platform to study the response of air quality along with aerosol chemical and physical properties to the strict emission control. Reduced anthropogenic emissions lowered the concentrations of primary pollutants and aerosol light absorption, but enhanced O₃ concentrations. Moreover, we found that meteorology was an important factor affecting on variations of air pollution. This did suggest that meteorological conditions should be considered when the control measures for improvement of air quality were made. Consequently, our study highlighted that how to get the balance among particulate matters (PM), NOx and O₃ along with meteorology will be an important scientific issue for co-improvement of PM and O₃ pollution in this urban city of China.

### Table 3

| Air pollutants | C_t | C_m | C_e |
|----------------|-----|-----|-----|
| BC (μg m⁻³)    | 1.1 | -0.54 (49%) | -0.56 (51%) |
| PM₂.₅ (μg m⁻³) | 33  | -12 (36%)    | -21 (64%)  |
| NO₂ (ppb)      | -9.1| -3.6 (40%)   | -5.5 (60%) |
| O₃ (ppb)       | 9.0 | -4 (-44%)    | 13 (144%)  |

*The values in the parentheses are the relative contributions of C_m or C_e to C_t, respectively.*

**Author statement**

Yu-Chi Lin: Writing – review & editing, Conceptualization Yan-Lin Zhang: Project administration Feng Xie: Methodology, Data curation, Formal analysis Mei-Yi Fan: Data curation Xiaoyan Liu: Data curation

**Declaration of competing interest**

The authors declare that they have no conflicting interest.
Acknowledgements

This study was financially supported by the Natural Scientific Foundation of China (No. 41977305), the Provincial Natural Science Foundation of Jiangsu (Grant No. BK20180040) and Jiangsu Innovation & Entrepreneurship Team. The associated data of this study can be downloaded online (https://doi.org/10.5281/zenodo.3963730).

Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.envpol.2021.116615.

References

Andreae, M.O., Crutzen, P.J., 1997. Atmospheric aerosols: biogeochemical sources and role in atmospheric chemistry. Science 276, 1052–1058. https://doi.org/10.1126/science.276.5315.1052.

Bao, R., Zhang, A., 2020. Does lockdown reduce air pollution? Evidence of 44 cities in northern China. Sci. Total Environ. 731, 139052. https://doi.org/10.1016/j.scitotenv.2020.139052.

Bauwens, M., Compernel, S., Stavroulakis, T., Müller, J.-F., van Gent, J., Eske, H., Levelt, P.F., van der A, R.V., Veldkamp, F., Vliegen, J.Y., Hu, J., Zehner, C., 2020. Impact of coronavirus outbreak on NO2 pollution assessed using TROPOMI and OMI observations. Geophys. Res. Lett. 47 https://doi.org/10.1029/2020GL087787.

Bond, T.C., Doherty, S.J., Fahey, D.W., Forster, P.M., Berntsen, T., Bennet, E., Dommen, J., Du, Q., Ghan, S., Kärcher, B., Koch, D., Kinne, S., Kondo, Y., Quinn, P.K., Sarofim, M.C., Schultz, M., Venkataraman, C., Zhang, H., Zhang, S., Bellouin, N., Ghan, D.,fd., Hopke, P.K., Jacobson, M.Z., Klimov, I., Lohmann, U., Schwarz, J.P., Shindell, D., Strowehm, T., Warren, S.G., Zender, C.S., 2013. Bonding the role of black carbon in the climate system: a scientific assessment. J. Geophys. Res. Atmos. 118, 5380–5525. https://doi.org/10.1002/jgrd.50371.

Bosch, C., Andersson, A., Kirillova, E.N., Budhavant, K., Tiwari, S., Praveen, P.S., Dumka, U.C., Kaskaoutis, D.G., Devera, P.C.S., Kumar, R., Kumar, S., Tiwari, S., Mihalopoulos, N., 2018. Year-long variability of fossil fuel and domestic wood burning black carbon component at a rural site in southern Delhi outburst region. Atmos. Chem. Phys. 18, 10583–10602. https://doi.org/10.5194/acp-18-10583-2018.

Chen, X., Wang, X., Wang, K., Wang, Z., Sun, J., Li, Z., Wang, L., Zhang, Y., Liang, S., Peng, Y., Zheng, J., Kang, H., 2019. Source apportionment of black carbon in different seasons in the northern suburban Nanjing, China. Atmos. Environ. 201, 190–200. https://doi.org/10.1016/j.atmosenv.2018.12.060.

Kirchsteetter, T.W., Novakov, T., Hobbs, P.V., 2004. Evidence that the spectral dependence of light absorption by aerosols is affected by organic aerosols. J. Geophys. Res. 109, D21208. https://doi.org/10.1029/2004JD004999.

Lack, D.A., Langridge, J.M., 2013. On the attribution of black and brown carbon light absorption using the Angström exponent. Atmos. Chem. Phys. 13, 10535–10543. https://doi.org/10.5194/acp-13-10535-2013.

Laskin, A., Laskin, J., Nizkorodov, S.K., 2015. Chemistry of atmospheric brown carbon. Chem. Rev. 115, 4335–4382. https://doi.org/10.1021/cr5006167.

Li, K, Jacobs, D.J., Liao, H., Shen, L., Zhang, Q., Bates, K.H., 2019. Anthropogenic drivers of 2013-2017 trends in surface ozone in the United States. Nat. Acad. Sci. Unit. States Am. 116, 422–427. https://doi.org/10.1073/pnas.1821681116.

Li, L., Qi, H., Huang, L., Wang, Q., Zhu, A., Xu, J., Li, Z., Li, H., Shi, L., Li, R., Azari, M., Wang, Y., Zhang, L., Khlystov, A., Dabdub, D., Chen, B., 2020. Air quality changes during the COVID-19 lockdown over the Yangtze Delta Region: an insight into the impact of human activity pattern changes on air pollution variation. Sci. Total Environ. 732, 139282. https://doi.org/10.1016/j.scitotenv.2020.139282.

Lin, C., Penner, J.E., Flanner, M.G., Sillman, S., Xu, L., Zhou, C., 2014. Radiative forcing of organic aerosol in the atmosphere and on snow: effects of SOA and brown carbon. J. Geophys. Res. Atmos. 119, 7453–7476. https://doi.org/10.1002/2013JD021186.

Li, X., Zhang, Y.-L., Peng, Y., Xu, L., Zhu, C., Cao, F., Zhai, X., Ma, M., Yang, C., Chang, Y., Huang, T., Xu, Z., Bao, M., Zhang, W., Fan, M., Li, X., 2019. Chemical and optical properties of carbonaceous aerosols in Nanjing, eastern China: regionally supported biogenic light absorption contributions. Atmos. Chem. Phys. 19, 11213–11233. https://doi.org/10.5194/acp-19-11213-2019.

Mousavi, A., Sowlat, M.H., Lovett, C., Rauber, M., Sridat, S., Boifi, R., Borgini, A., De Marco, C., Ruprecht, A.A., Sioutas, C., 2019. Source apportionment of black carbon (BC) from fossil fuel and biomass burning in metropolitan Milan, Italy. Atmos. Environ. 203, 252–261. https://doi.org/10.1016/j.atmosenv.2019.02.009.

Ni, H., Huang, R.-J., Cao, J., Zhang, T., Wang, M., Meijer, H.A.J., Dusek, U., 2018. Source apportionment of carbonaceous aerosols in Xian, China: insights from a full year of measurements of radiocarbon and the stable isotope 13C. Atmos. Chem. Phys. 18, 16363–16383. https://doi.org/10.5194/acp-18-16363-2018.

Nozère, B., Dziedzic, P., Córdova, A., 2007. Formation of secondary light-absorbing “fuficike” oligomers: a common process in aqueous and iconic atmospheric water. Geophys. Res. Lett. 34, L12812. https://doi.org/10.1029/2007GL031300.

Olson, M.R., Garcia, M.V., Robinson, M.A., Rooy, P.V., Dieterenberger, M.A., Bergin, M., Schaer, J., 2015. Investigation of black and brown carbon multiple-wavelength light absorption from biomass and fossil fuel combustion source emissions. J. Geophys. Res. Atmos. 120, 6682–6697. https://doi.org/10.1002/2014JD022970.

Ramanathan, V., Carmichael, G., 2008. Global and regional climate changes due to black carbon. Nat. Geosci. 1, 227–228. 10.1038/ngeo156.

Rice, T.D., Andreae, M.O., 2014. Contribution of wood burning to PM10 in London. Atmos. Environ. 87, 89–94. https://doi.org/10.1016/j.atmosenv.2013.12.037.

Ugolini, L., Cappelletti, B., Capri, L., Longo, M., Ermolli, I., Rizzoli, P., 2013. Monitoring of black carbon and elemental carbon in the South Asian outburst region. Atmos. Chem. Phys. 13, 2391–2405. https://doi.org/10.5194/acp-13-2391-2013.

Vega, S., Mihalopoulos, N., 2019. Optical properties of carbonaceous aerosols in Nanjing, eastern China: regionally supported biogenic light absorption contributions. Atmos. Chem. Phys. 19, 11213–11233. https://doi.org/10.5194/acp-19-11213-2019.
Wang, C., Horby, P.W., Hayden, F.G., Gao, G.F., 2020a. A novel coronavirus outbreak of global health concern. Lancet 395, 470–473. https://doi.org/10.1016/S0140-6736(20)30185-9.

Wang, Q., Ye, J., Wang, Y., Zhang, T., Ran, W., Wu, Y., Tian, J., Li, L., Zhou, Y., Ho, S.S.H., Deng, B., Zhang, Q., Zhang, R., Chen, Y., Zhu, C., Cao, J. 2019. Winter optical properties of primary and secondary brown carbon at a regional site in the Northern China Plain. Environ. Sci. Technol. 53, 12389–12397. https://doi.org/10.1021/acs.est.9b03406.

Wang, Y., Yuan, Y., Wang, Q., Liu, C.G., Zhi, Q., Cao, J. 2020b. Changes air quality related to the control of coronavirus in China: implications for traffic and industrial emissions. Sci. Total Environ. 731, 139133. https://doi.org/10.1016/j.scitotenv.2020.139133.

Watson, J.G., 2002. Visibility: science and regulation. J. Air Waste Manag. Assoc. 52, 626–713. https://doi.org/10.1080/10473289.2002.10470813.

Wu, C., Yu, J.Z. 2016. Determination of primary combustion source organic carbon-to-elemental carbon (OC/EC) ratio using ambient OC and EC measurements: secondary OC-EC correlation minimization method. Atmos. Chem. Phys. 16, 5453–5465. https://doi.org/10.5194/acp-16-5453-2016.

Wu, F., Zhao, S., Yu, B., Chen, Y., Wang, W., Song, Z., Hu, Y., Tao, Z., Tian, J., Pei, Y., Yuan, M., Zhang, Y., Dai, F., Liu, Y., Wang, Q., Zheng, J., Xu, L., Holmes, E.C., Zhang, Y. 2020. A new coronavirus associated with human respiratory disease in China. Nature 579, 265–269. https://doi.org/10.1038/s41586-020-2008-3.

Yan, C., Zheng, M., Bosch, C., Andersson, A., Desyaterik, Y., Sullivan, A.P., Collett, J.L., Zao, B., Wang, S., He, K., Gustafsson, O. 2017. Important fossil sources contribution to brown carbon in Beijing during winter. Sci. Rep. 7, 43182. https://doi.org/10.1038/srep43182.

Yang, M., Howell, S.G., Zhuang, J., Huebert, B.J., 2009. Attribution of aerosol light absorption to black carbon, brown carbon and dust in China—interpretations of atmospheric measurements during EAST-AIRE. Atmos. Chem. Phys. 9, 2033–2050. https://doi.org/10.5194/acp-9-2033-2009.

Yuan, W., Huang, R.-J., Yang, L., Guo, J., Chen, Z., Duan, J., Wang, M., Wang, T., Ni, H., Han, Y., Li, Y., Chen, Q., Chen, Y., Hoffman, T., O’Dowd, C. 2019. Characterization of the light absorbing properties, chromophores composition and source of brown carbon aerosol in Xian, Northwest China. Atmos. Chem. Phys. 20, 5129–5144. https://doi.org/10.5194/acp-20-5129-2020.

Zhang, Q., Shen, Z., Zhang, L., Zeng, Y., Ning, Z., Zhang, T., Lei, Y., Li, G., Sun, J., Westerdahl, D., Xu, H., Cao, J. 2020. Investigation of primary and secondary particulate brown carbon in two Chinese cities: Xian and Hong Kong in winter. Environ. Sci. Technol. 54, 3803–3813. https://doi.org/10.1021/acs.est.9b05332.

Zhang, Y., Ren, H., Sun, Y., Cao, F., Chow, J., Liu, S., Lee, X., Agrios, K., Kawamura, K., Liu, D., Ren, L., Du, W., Wang, Z., Prevot, A.S.H., Szidat, S., Fu, P. 2017. High contribution of nonfossil source to submicrometer organic aerosols in Beijing. Environ. Sci. Technol. 51, 7842–7852. https://doi.org/10.1021/acs.est.7b01517.

Zhao, B., Wang, P., Ma, J.Z., Zhu, S., Pozzer, A., Li, W. 2012. A high-resolution emission inventory of primary pollutants for the Huabei region, China. Atmos. Chem. Phys. 12, 481–501. https://doi.org/10.5194/acp-12-481-2012.