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Decrease of atmospheric black carbon and CO\textsubscript{2} concentrations due to COVID-19 lockdown at the Mt. Waliguan WMO/GAW baseline station in China

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\section*{ABSTRACT}

The Coronavirus Disease 2019 (COVID-19) lockdown policy reduced anthropogenic emissions and impacted the atmospheric chemical characteristics in Chinese urban cities. However, rare studies were conducted at the high mountain site. In this work, in-situ measurements of light absorption by carbonaceous aerosols and carbon dioxide (CO\textsubscript{2}) concentrations were conducted at Waliguan (WLG) over the northeastern Tibetan Plateau of China from January 3 to March 30, 2020. The data was employed to explore the influence of the COVID-19 lockdown on atmospheric chemistry in the background-free troposphere. During the sampling period, the light absorption near-infrared (\textasciitilde 720 nm) was mainly contributed by BC (\textasciitilde 72\%), however, BC and brown carbon (BrC) contributed equally to light absorption in the short wavelength (\textasciitilde 350 nm). The average BC concentrations in the pre-, during and post-lockdown were 0.28 \pm 0.25, 0.18 \pm 0.16, and 0.28 \pm 0.20 \mu g \text{m}^{-3}, respectively, which decreased by approximately 35\% during the lockdown period. Meanwhile, CO\textsubscript{2} also showed slight decreases during the lockdown period. The declined BC was profoundly attributed to the reduced emissions (\textasciitilde 86\%), especially for the combustion of fossil fuels. Moreover, the declined light absorption of BC, primary and secondary BrC decreased the solar energy absorbance by 35, 15, and 14\%, respectively. The concentration weighted trajectories (CWT) analysis suggested that the decreased BC and CO\textsubscript{2} at WLG were exclusively associated with the emission reduction in the eastern region of WLG. Our results highlighted that the reduced anthropogenic emissions attributed to the lockdown in the urban cities did impact the atmospheric chemistry in the free troposphere of the Tibetan Plateau.

\section*{1. Introduction}

Black Carbon (BC, or “soot”) particles in the atmosphere are mainly produced from incomplete combustion of carbonaceous fuels, e.g. wood-burning, fossil fuels, and biofuels (Bond et al., 2013). After carbon dioxide (CO\textsubscript{2}), BC has been considered the second greatest contributor to global warming (Bond et al., 2013). The direct radiative forcing effect of BC aerosols is 0.24 \pm 0.14 W m\textsuperscript{-2} (Liao and Chang, 2014). BC has the ability to strongly absorb solar radiation and affect the Earth’s radiation budget through direct, indirect, and semi-direct effects (Bond et al., 2013; Goel et al., 2021). Studies have shown the heating effect in the high-altitude Himalayan-Tibetan region was mainly attributed to BC (Lau et al., 2010). The deposition of particles particularly BC over glaciers of the Tibetan Plateau region of China contributes to their

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premature thawing and fragmentation as it produces the albedo effect that accelerates snow melting (Menon et al., 2010; Y. Zhang et al., 2020). CO₂ is the main greenhouse gas responsible for radiative forcing of up to 1.82 ± 0.19 W m⁻², which was 9 times higher than that of BC. Energy consumption (including fossil fuels) has increased sharply in recent years as a result of urbanization and modern lifestyle changes. Fossil fuels are responsible for 85% of CO₂ emissions and 64% of total greenhouse gas emissions (Razzaq et al., 2020)

Coronavirus Disease 2019 (COVID-19) outbreak began worldwide in January 2020. China was the first country to report COVID-19 and implemented a series of effective control measures (called COVID-19 lockdown thereafter) to prevent the spread of the virus. The measures included stay-at-home orders, local and international travel bans, control on commercial and human activities (Tian et al., 2020). These control measures resulted in strict reduction of anthropogenic emissions and changes in concentrations of air pollutants. A previous study has suggested that BC mass concentrations during the COVID-lockdown periods were reduced by 44% in Hangzhou of China (Xu et al., 2020). During the lockdown, the concentrations of PM₂.₅ and nitrogen dioxide (NO₂) in North China decreased by ~ 35% and 60%, respectively (Shi and Brasseur, 2020). In eastern China, COVID-19 lockdown resulted in a decrease of PM₂.₅ by 12.5% (Q. Zhang et al., 2021). Due to the COVID-19 lockdown, the substantial decreases of BC emissions mainly attributed to combustion sources were found in eastern and northern China with a decreasing rate of 48%-70% compared with those in the pre-lockdown (Jia et al., 2021). The reduced BC emissions resulted in significant decreases of BC concentration in the suburbs of Nanjing (41%) and Shandong Province (32%) (Y.-C. Lin et al., 2021). In addition to BC, a significant decrease of CO₂ concentration was also observed in Chinese urban and remote areas. For instance, Wu et al. (2021) found that CO₂ in Xi’an decreased by 7.5% during the lockdown period (Wu et al., 2021). In the remote region, Liu et al. (2021a, b) found that the apparent decrease of BC concentration during COVID-19 in Tibet was attributed to huge reduced anthropogenic emissions in South Asia.

Previous many studies have focused on the investigation of impacts of COVID-19 lockdown on concentrations of air pollutants and changes of air quality (Ambade et al., 2021; Evangeliou et al., 2021; Goel et al., 2021). However, most of these studies were conducted in Chinese urban cities, only one research was carried out at the Tibetan background site (Liu et al., 2021a, b). In this work, we continuously monitored the light absorption by carbonaceous aerosols at the Waliguan (WLG) station from 2020 January 3 to March 31 when covered the COVID-19 lockdown period (from 3 February to 19 February). The regularly monitored CO₂ data at this mountainous site were also acquired. Using these data sets, we characterized the light absorption by BC and brown carbon (BrC) at WLG. The absolute concentrations and source apportionments (biomass burning vs. fossil fuel) of BC were also estimated. Compared to those in the pre-lockdown, the impacts of reduced anthropogenic emissions during the COVID-19 lockdown on light absorption, sources, and solar energy absorbance by carbonaceous aerosols and the CO₂ concentrations at WLG were also discussed. The results would provide a hint to understand the impacts of anthropogenic emissions in the downhill regions on the atmospheric chemistry in the free troposphere in Qinghai Plateau.

2. Methodology

2.1. Sampling site

The sampling site Waliguan (WLG, 36°17’N, 100°54’E, 3816 m a.s.l.) is located in the northeastern part of the Qinghai-Tibet Plateau, China (Fig. 1). It is one of the Global Atmospheric Watch (GAW) baseline stations of the World Meteorological Organization (WMO) and the only one in the hinterland of the Eurasian continent. The sampling site is located on an isolated hilltop with no surrounding mountains (Ma et al., 2020). There is no industrial emission within the 30 km circle of the sampling site, and the two main potential source regions of pollutants are Xining city in the north (90 km away) and Lanzhou city in the east (260 km away). Therefore there is no local source of pollution that can directly influence the sampling site. The continuous real-time measurements of BC light absorption were conducted from 3 January to 31 March in 2020 using a seven-channel Magee® AE33 aethalometer

Fig. 1. Relative locations of the sampling site (colored by altitude from NASA). Red circle is the sampling site and blue dots are the large cities surrounded by the sampling site. (For interpretation of the references to color in this figure legend, the reader is referred to the Web version of this article.)
installed at the rooftop of a two-story building, which was at a height of 5 m from the ground. In addition, the concentrations of PM$_{2.5}$, CO$_2$, and carbon monoxide (CO) were obtained from the WLG monitoring station. Meteorological data such as temperature, humidity, wind direction, and wind speed were obtained from the automatic weather station. The height of the boundary layer was obtained by analyzing the data through the Global Data Assimilation System (GDAS) from National Centre for Environmental Planning (NCEP), and the time and frequency were 3 h.

2.2. Light absorption measurement

In this work, an Aethalometer (model AE33, Magee Scientific) was used to monitor BC light absorption at WLG. The instrument measures light attenuation at seven wavelengths of 370, 470, 520, 590, 660, 880, and 950 nm, which is linearly proportional to the BC mass deposited over the filter tape (Drinovec et al., 2015; Hansen et al., 1984; Helin et al., 2018; Q. Wang et al., 2019c). 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 then measured with a time resolution of 1 min. The BC concentrations can be calculated by ATN and the mass absorption cross-section (MAC) of 7.77 m$^2$ g$^{-1}$ since the light absorption at this wavelength with other species is insignificant (Drinovec et al., 2015). The BC concentrations can be calculated as:

$$BC_{\text{measured}} = \frac{S \cdot ATN}{\sigma_{BC} \cdot F_{\text{inlet}} \cdot \alpha}$$  \hspace{1cm} (1)

where S is the aerosol loading area (m$^2$) and $F_{\text{inlet}}$ is the real-time sampling flow rate (L min$^{-1}$), C is the correction factor for multiple scattering effects in the filter strip (2.14 in AE33) and $\alpha$ is the mass absorption cross-section (m$^2$ g$^{-1}$). However, with BC concentrations in the sampling process, there was a loading effect leading to inaccurate sampling results (Drinovec et al., 2015) and AE33 two-point measurements were applied to solve this problem.

$$BC = BC_{\text{measured}}/(1 - k \cdot ATN)$$  \hspace{1cm} (2)

where $BC_{\text{measured}}$ is the BC concentration calculated from a single point of movement and $k$ is the load correction factor, which can be obtained by fitting two different flow measurements of BC to ATN.

2.3. Primary and secondary BrC absorption

BC is the main light-absorbing substance in the visible band, with Ångström absorption exponent (AAE) of 1.0. Another light-absorbing substance in the visible band is BrC, which has a significant absorption peak around 365 nm, and the AAE of incompletely burned biomass BrC in the visible band is 1–3, reaching 3–7 after conversion to secondary BrC (Yan et al., 2018). The aerosol light absorption is dependent on the different wavelengths (Laskin et al., 2015):

$$b_{abs}(\lambda) = k \lambda^{-\text{AAE}}$$  \hspace{1cm} (3)

where k is a constant which is related to the aerosol mass concentration. Without consideration of dust particles to light absorption, $b_{abs}(\lambda)$ can then be divided into $b_{abs,BC}$, $b_{abs,BrC}$, and $b_{abs,BrC_{sec}}$ (Q. Wang et al., 2019b, c)

$$b_{abs}(\lambda) = b_{abs,BC}(\lambda) + b_{abs,BrC_{sec}}(\lambda) + b_{abs,BrC}(\lambda)$$  \hspace{1cm} (4)

The light absorption at the wavelength of 370, 470, 520, 590, and 660 nm are considered in Eq. (7). Assuming the absorption of BrC at the wavelengths of 880 nm is only induced by BC, the light absorption of BC at the wavelengths of 370, 470, 520, 590, and 660 nm can then be calculated as:

$$b_{abs,BC}(\lambda) = b_{abs}(880) \times \left( \frac{\lambda}{880} \right)^{-\text{AAE}_{\text{BC}}}$$  \hspace{1cm} (5)

The $b_{abs,BrC}$ can be calculated as (Q. Wang et al., 2019c):

$$b_{abs,BrC_{sec}}(\lambda) = b_{abs}(\lambda) - \left( b_{abs}(880) \right)_{\text{pri}} \times [\text{BC}]$$  \hspace{1cm} (6)

where $b_{abs}(880)/[\text{BC}]_{\text{pri}}$ is the $b_{abs}(\lambda)$-to-BC ratio in primary emission sources at wavelengths of 370, 470, 520, 590 and 660 nm with a unit of m$^2$ g$^{-1}$. [BC] is the concentration of particulate BC (μg m$^{-3}$), which was retrieved from the relationship between the ATN and mass absorption cross-section (MAC) at a wavelength of 880 nm. The ratio of $b_{abs}(880)/[\text{BC}]_{\text{pri}}$ varied with different emission sources and therefore we used a minimum $R^2$-squared (MRS) approach to obtain the appropriate values of $b_{abs}(\lambda)/[\text{BC}]_{\text{pri}}$.

After obtaining the $b_{abs,BrC_{sec}}$, we calculated the light absorption of primary $b_{abs}$, $b_{abs,BrC}$ as:

$$b_{abs,BrC_{sec}}(\lambda) = b_{abs}(\lambda) - b_{abs,BrC_{sec}}(\lambda) - b_{abs,BC}(\lambda)$$  \hspace{1cm} (7)

2.4. Partition of fossil fuel and biomass burning to BC

The Aethalometer has the ability to quantify the relative proportion of BC from fossil fuel (FF) and biomass burning (BB) emissions (J. Sandradewi et al., 2008a, b). Here, the aerosol light absorption at wavelengths of 470 and 950 nm was used to quantify the relative contributions of BB and FF to particulate BC. The equations relating the light absorption ($b_{abs}(\lambda)$), wavelengths (470 and 950 nm), and absorption Ångström exponent (AAE) were expressed as (Sandradewi et al., 2008a, b):

$$b_{abs}(470\text{nm})_{BB} = \left( \frac{470}{950} \right)^{-\text{AAE}_{\text{BB}}}$$  \hspace{1cm} (8)

$$b_{abs}(950\text{nm})_{BB} = \left( \frac{470}{950} \right)^{-\text{AAE}_{\text{FF}}}$$  \hspace{1cm} (9)

$$b_{abs}(\lambda) = b_{abs,FF}(\lambda) + b_{abs,BB}(\lambda)$$  \hspace{1cm} (10)

The AAE$_{\text{BB}}$ values of 1.8–2.2 and AAE$_{\text{FF}}$ of 0.9–1.1 have been widely used in the Aethalometer model (Díaz Resquin et al., 2018; Drinovec et al., 2015; Fuller et al., 2014; Herich et al., 2011; Titos et al., 2017). Here, the AAE$_{\text{BB}}$ and AAE$_{\text{FF}}$ values were assumed to be 2.0 and 1.0, respectively, for the estimations (J. Sandradewi et al., 2008a, b).

Combining the Eqs. (3)–(5), we can obtain the proportion of BB (%) contributed by BB activities (J. Sandradewi et al., 2008a, b):

$$BB(\%) = \frac{b_{abs}(950\text{nm})_{BB}}{b_{abs}(950\text{nm})} \times 100$$  \hspace{1cm} (11)

The absolute concentration of BB was calculated as:

$$BC_{BB} = BC(880\text{nm}) \cdot BB$$  \hspace{1cm} (12)

After obtaining the absolute concentration of BC$_{BB}$, the absolute BC$_{FF}$ concentrations were obtained.

$$BC_{FF} = BC(880\text{nm}) - BC_{BB}$$  \hspace{1cm} (13)

2.5. Calculation of solar energy absorbance

We calculated the total solar energy absorbance ($E_{\text{Total}}$) of carbonaceous aerosols (using wavelength range from 300 to 950 nm) from the summation of those by BC (E$_{BC}$) and brown carbon (E$_{BrC}$) (Liu et al., 2019):

$$E_{\text{Total}} = E_{BC} + E_{BrC}$$  \hspace{1cm} (14)
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 at WLG during the sampling period.

| Parameters       | 370 | 470 | 520 | 590 | 660 | 880 | 950 |
|------------------|-----|-----|-----|-----|-----|-----|-----|
| $b_{abs,total}$  |     |     |     |     |     |     |     |
| $b_{abs,BC}$     | 8.76 ± 6.76 | 5.28 ± 4.21 | 4.15 ± 3.40 | 3.37 ± 2.82 | 2.81 ± 2.36 | 2.03 ± 1.67 | 2.03 ± 1.67 | 1.88 ± 1.54 |
| 55%              | 4.83 ± 3.96 | 3.80 ± 3.11 | 3.43 ± 2.81 | 3.03 ± 2.48 | 2.71 ± 2.22 | 2.03 ± 1.67 | 1.88 ± 1.55 |
| $b_{abs,BrC-pri}$| 2.48 ± 2.78 | 0.80 ± 1.17 | 0.26 ± 0.67 | 0.04 ± 0.43 | 0 ± 0.27 | 1% | 1% | 100% |
| 28%              | 1.47 ± 1.75 | 0.68 ± 0.54 | 0.46 ± 0.31 | 0.29 ± 0.19 | 0.26 ± 0.17 | 9% | 3% | 100% |

The Aethalometer AE33 directly measured the aerosol light absorption of PM$_{2.5}$ at seven wavelengths. Table 1 lists the aerosol light absorption of BC, primary and secondary BrC through the different wavelengths at WLG during the sampling period. The results showed that the average light absorption at 370, 470, 520, 590, 660, 880 and 950 nm wavelengths were 8.76 ± 6.76, 5.28 ± 4.21, 4.15 ± 3.40, 3.37 ± 2.82, 2.81 ± 2.36, 2.03 ± 1.67, and 1.88 ± 1.54 Mm$^{-1}$, respectively. BC was the dominant species of light-absorbing carbonaceous aerosols with a relative contribution to the total light absorption from 55% ($\lambda = 370$ nm) to 100% ($\lambda = 880$ and 950 nm), followed by primary BrC (from 1% at $\lambda = 660$ nm to 28% at $\lambda = 370$ nm) and secondary BrC. The fraction of BrC to light absorption in the plateau was 20% higher than in the suburbs of Nanjing (around 30% in the suburbs of Nanjing) (Y.-C. Lin et al., 2021), suggesting that the lower contribution of BC light absorption in this mountain site.

The BC light absorption increased with increases of the light wavelengths whereas the absorption of primary and secondary BrC decreased with the increasing light wavelengths. The average light absorption by BC, BrC$_{pri}$, and BrC$_{sec}$ at the different wavelengths is shown in Fig. 5I. The average AAE value for the whole period was 1.80 ± 0.11 and was 1.3–1.6 times lower than the urban areas (Jing et al., 2019; Y.-C. Lin et al., 2021). A larger AAE indicated a stronger light absorption by BrC at WLG. Before the lockdown, the AAE value was 1.76 ± 0.09, which was lower than that (2.06 ± 0.14) during the lockdown period. After the lockdown, the AAE values rose up to 1.91. The lower AAE values during the lockdown period suggested that a shift of the main source to the light absorption from FF to BB and the presence of a decrease of fossil fuel emission sources at WLG during the lockdown.

3. Results and discussion

3.1. Absorption characteristics in visible wavelengths

The Aethalometer AE33 directly measured the aerosol light absorption of PM$_{2.5}$ at seven wavelengths. Table 1 lists the aerosol light absorption of BC, primary and secondary BrC through the different wavelengths at WLG during the sampling period. The results showed that the average light absorption at 370, 470, 520, 590, 660, 880 and 950 nm wavelengths were 8.76 ± 6.76, 5.28 ± 4.21, 4.15 ± 3.40, 3.37 ± 2.82, 2.81 ± 2.36, 2.03 ± 1.67, and 1.88 ± 1.54 Mm$^{-1}$, respectively. BC was the dominant species of light-absorbing carbonaceous aerosols with a relative contribution to the total light absorption from 55% ($\lambda = 370$ nm) to 100% ($\lambda = 880$ and 950 nm), followed by primary BrC (from 1% at $\lambda = 660$ nm to 28% at $\lambda = 370$ nm) and secondary BrC. The fraction of BrC to light absorption in the plateau was 20% higher than in the suburbs of Nanjing (around 30% in the suburbs of Nanjing) (Y.-C. Lin et al., 2021), suggesting that the lower contribution of BC light absorption in this mountain site.

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significant decreases of BC concentrations during the lockdown period were found (Fig. 2). On average, the BC concentration was $0.18 \pm 0.16 \, \mu g \, m^{-3}$ during the lockdown, which decreased by 35% compared with those in the pre- ($0.28 \pm 0.25 \, \mu g \, m^{-3}$) and post-lockdown ($0.28 \pm 0.20 \, \mu g \, m^{-3}$). Meanwhile, the CO$_2$ concentrations decreased from 414.4 ppm in the pre-lockdown to 413.4 ppm in the lockdown (at a 95% significance level). A good correlation between CO$_2$ and BC with $R^2$ value ranging from 0.71 to 0.86 ($p < 0.05$) (Fig.S2), indicating that CO$_2$ was likely to be mainly constrained by anthropogenic emissions (combustion sources) rather than natural emissions during the sampling period. The CO concentrations also decreased (from $0.17 \pm 0.01$ ppm in the pre-lockdown to $0.13 \pm 0.05$ ppm in the lockdown) (at a 95% significance level). CO is a product from incomplete combustion, such as vehicle emissions and industrial processes. Consequently, the declined CO concentrations also reflected the reduced emissions of traffic and industrial emissions in the COVID-19 lockdown. The PM$_{2.5}$ concentrations also decreased by 39% due to lockdown policy ($6.3 \pm 5.2 \, \mu g \, m^{-3}$ in the lockdown vs. $10.2 \pm 9.2 \, \mu g \, m^{-3}$ in the pre-lockdown), while ozone (O$_3$) concentration did not change significantly during the pre- and lockdown period. During the same period, BC concentrations decreased by 41% and 44% in Nanjing and Hangzhou, respectively, while PM$_{2.5}$ concentrations declined 57% (Nanjing) (Y.-C. F. Xie et al. 2021, Environmental Research 211 (2022) 112984.
The diurnal variations of BC concentrations at WLG are illustrated in Fig. 3. As shown, the similar diurnal patterns were observed in the pre- and post-lockdown, the lower BC, CO, and CO\textsubscript{2} concentrations were observed in the lockdown, suggesting that the reduced anthropogenic emissions also decreased the BC, CO, and CO\textsubscript{2} concentrations in the free troposphere over the Qinghai Plateau.

### 3.3. Influence of reduced emission on BC mass concentrations

Since the strict reduced anthropogenic emissions were observed, the emission sources of BC at WLG might change during the lockdown period. In this study, we used the Aethalometer model to partition the relative contributions of BB and FF to BC at WLG and the results are shown in Fig. 4a. As shown, FF contributed a major part of BC at WLG during the sampling period, with a relative contribution of 58 ± 12%. However, the partitioned sources were different during the distinct periods. Before and after the lockdown, the average contributions of FF to BC were both approximately 62 ± 11%. On the contrary, the contribution of FF to BC dropped to 52 ± 14% in the lockdown. The FF to BC decreased by 10% during the lockdown period (p < 0.05). This decreasing rate was in line with those in Nanjing and Hangzhou (Y.-C. Lin et al., 2021; Xu et al., 2020). Although the uncertainty of FF to BC (13.9%, see in Supporting Information S1) was existed in the estimation, our results implied that the shutdown of vehicle and industries significantly decreased the relative contribution of fossil fuel to BC aerosols at this mountain site. In the atmosphere, BC, CO, and CO\textsubscript{2} are mainly produced from combustion sources and BC/CO ratios can be used to identify the potential sources of BC. Consequently, we utilized \( \Delta \text{BC}/\Delta \text{CO}_2 \) (the slope of BC-CO\textsubscript{2}) and \( \Delta \text{BC}/\Delta \text{CO} \) (the slope of BC-CO) to determine the potential sources of BC; the results are plotted in Fig. 4b and c. Also, we listed \( \Delta \text{BC}/\Delta \text{CO} \) and \( \Delta \text{BC}/\Delta \text{CO}_2 \) ratios of various combustion sources in Table 3 (Liñán-Abanto et al., 2021; Verma et al., 2020).

### Table 2

The mean values of the concentration in various air pollutants, meteorological parameters and light absorption together with solar energy absorbance before, during, and after the lockdown.

| Species/parameters | before  | lockdown | after |
|--------------------|---------|----------|-------|
| \( \text{PM}_{2.5} \) (\( \mu \text{g m}^{-3} \)) | 10.20 ± 9.16 | 6.26 ± 5.23 | 10.28 ± 7.50 |
| BC (\( \mu \text{g m}^{-3} \)) | 0.28 ± 0.25 | 0.18 ± 0.16 | 0.28 ± 0.20 |
| CO (ppm) | 0.17 ± 0.01 | 0.13 ± 0.05 | 0.14 ± 0.05 |
| CO\textsubscript{2} (ppm) | 414.42 ± 3.84 | 413.39 ± 1.91 | 414.66 ± 2.56 |
| \( \text{O}_3 \) (ppb) | 46.46 ± 3.90 | 46.16 ± 2.65 | 49.60 ± 6.03 |
| T (°C) | 11.09 ± 3.5 | 9.46 ± 3.03 | −4.72 ± 4.05 |
| RH (%) | 50.77 ± 21.24 | 22.93 ± 12.75 | 41.22 ± 25.43 |
| WS (m s\textsuperscript{-1}) | 4.01 ± 2.45 | 4.67 ± 3.24 | 4.78 ± 2.97 |
| BLH (m) | 223 ± 334 | 385 ± 634 | 564 ± 804 |
| BC absorption (Mm\textsuperscript{-1}) | 5.09 ± 4.57 | 3.26 ± 2.83 | 5.26 ± 3.71 |
| Primary BrC absorption (Mm\textsuperscript{-1}) | 2.25 ± 2.54 | 2.17 ± 2.48 | 2.78 ± 3.08 |
| Secondary BrC absorption (Mm\textsuperscript{-1}) | 1.46 ± 1.23 | 1.20 ± 1.06 | 1.56 ± 2.24 |
| \( \text{E}_{\text{BC}} (\text{W m}^{-2}) \) | 2.33 ± 2.08 | 1.50 ± 1.29 | 2.41 ± 1.68 |
| \( \text{E}_{\text{BC-pret}} (\text{W m}^{-2}) \) | 0.38 ± 0.58 | 0.32 ± 0.56 | 0.42 ± 0.63 |
| \( \text{E}_{\text{BC-sect}} (\text{W m}^{-2}) \) | 0.37 ± 0.22 | 0.32 ± 0.21 | 0.38 ± 0.37 |
| \( \text{E}_{\text{total}} (\text{W m}^{-2}) \) | 3.07 ± 2.66 | 2.14 ± 1.82 | 3.21 ± 2.25 |

Lin et al., 2021; Xu et al., 2020). CO\textsubscript{2} decreased by approximately 36 ppm (7.5%) in Xi’an, with 484.5 ± 21.4 ppm and 456.1 ± 9.1 ppm before and during the lockdown, respectively (Wu et al., 2021). These findings demonstrated that the substantial decreases in the concentrations of air pollutants during the lockdown period were observed not only at urban sites, but also in the free troposphere over continental China.

The diurnal variations of BC concentrations at WLG are illustrated in Fig. 3. As shown, the similar diurnal patterns were observed in the pre-, during- and post-COVID-19 lockdown. The diel cycles of BC concentrations at WLG were in agreement with those in other background mountainous sites (Y. C. Lin et al., 2011; Y. C. Lin et al., 2013), where the up- and down-slope winds played an important role for diurnal variations of air pollutants. Typically, BC showed relative flat values from the nighttime to early morning hours and then decreased after sunrise. After reaching a minimum value at 7:00 or 8:00 LT (local time), BC concentrations began to increase and peaked around noontime and then decreased due to the well-developed boundary layer. After sunrise, the boundary layer height (BLH) decreased, enhancing the BC concentrations. On the other hand, both CO and CO\textsubscript{2} showed the same diurnal patterns as BC. This was expected since the sources of CO, CO\textsubscript{2} and BC were similar and their concentrations at WLG were influenced by up- and down-slopes as well as BLH (García-Franco et al., 2018; Liñán-Abanto et al., 2021; Xiaolin Zhang et al., 2013). Compared to those in the pre- and post-lockdown, the lower BC, CO, and CO\textsubscript{2} concentrations were observed in the lockdown, suggesting that the reduced anthropogenic emissions also decreased the BC, CO, and CO\textsubscript{2} concentrations in the free troposphere over the Qinghai Plateau.

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**Fig. 3.** Diurnal cycles of BC, CO\textsubscript{2}, CO, and BLH in WLG.
During the sampling period, $\Delta$BC/$\Delta$CO$_2$ and $\Delta$BC/$\Delta$CO were $0.063 \pm 0.0007 \, \mu g \, m^{-3} \, ppm^{-1}$ ($R^2 = 0.79, p < 0.05$) and $0.0027 \pm 0.0001 \, \mu g \, m^{-3} \, ppb^{-1}$ ($R^2 = 0.78, p < 0.05$) respectively, suggesting that the sources of BC at WLG are transmitted gasoline or diesel and domestic coal combustion. Moreover, the values of both $\Delta$BC/$\Delta$CO$_2$ and $\Delta$BC/$\Delta$CO increased during the lockdown period, indicating the increases of the contributions of domestic coal and biofuels or biomass burning to BC and a decrease of vehicle diesel and gasoline or industry coals to BC. This implied that the reduced anthropogenic emissions over intensive-population regions did impact the atmospheric chemical compositions at the WLG station.

Furthermore, we calculated the absolute concentrations of BC from BB ($BC_{BB}$) and FF ($BC_{FF}$) by BC masses multiplying the fractions of BC contributed by BB and FF, respectively. Fig. 5 illustrates the diurnal patterns of concentrations in $BC_{BB}$ and $BC_{FF}$ at WLG in the pre-, during, and post-lockdown. The result showed that the diel cycles of $BC_{BB}$ and $BC_{FF}$ were very similar. From the nighttime to early morning on the next day, $BC_{BB}$ and $BC_{FF}$ kept relative stable concentrations and then decreased from the early morning, reaching minimum values from 6:00 LT to 8:00 LT. Subsequently, $BC_{BB}$ and $BC_{FF}$ concentrations increased and peaked around noontime, which were influenced by both meteorological conditions and emissions sources. $BC_{BB}$ and $BC_{FF}$ has the

**Table 3**

The ratios of BC/CO ($\mu g \, C \, m^{-3} \, ppb^{-1}$) and BC/CO$_2$ ($\mu g \, C \, m^{-3} \, ppm^{-1}$) derived from various emission sources (grams of pollutant evolved per kilogram of fuel burned).

| Source             | BC/CO | BC/CO$_2$ |
|--------------------|-------|-----------|
| Biomass burning    | 0.0056–0.016 | 0.53–1.10 |
| Transport          | 0.0013–0.055 | 0.15      |
| Industry           | 0.0031–0.0115 | 0.06–0.11 |
| Domestic           | 0.0017–0.0182 | 0.04–0.74 |
| Biofuels           | 0.0007–0.0266 | 0.69–1.75 |

a (Dhammapala et al., 2007).
b (Cao et al., 2008).
c (Andreae and Merlet, 2001).
d (Streets et al., 2003).
e (Dickerson et al., 2002).
f (Boucher and Reddy, 2008).
g (Sánchez-Coyollo et al., 2009).
h (Westerdahl et al., 2009).
similar diurnal cycles with BC, which due to the active of local residents (cooking and heating using yak’s dry dung) and tourists (traffic and other biofuel burnings) in some research (Zhang et al., 2017). The enhancements of BBFF and BBFF were attributed to the up-slope wind, which brought air pollution from downhill areas (Y. C. Lin et al., 2011). After noontime, the BLH developed well and resulted in declined BCBB and BCFF concentrations. After sunset, the decreases of BLH increased the BCBB and BCFF concentrations. The average BCBB concentration in the lockdown was 0.09 ± 0.08 μg m⁻³, which was similar to those (0.10 ± 0.08 and 0.11 ± 0.08) in the pre- and post-lockdown, indicating that the BB activities were still operated during the lockdown period over China. This might be true since BB in winter was regularly used for residential heating that was not prohibited for lockdown policy (Xu et al., 2020). On the contrary, the BCFF concentrations reduced by 48% compared with those before and after the lockdown period, suggesting that the strict reduced anthropogenic emissions via the COVID-19 lockdown did impact the atmospheric chemical compositions at WLG, which was representative of the free troposphere in northern China.

3.4. Influence of reduced emission ratio on BC mass concentrations

The reduced anthropogenic emissions via lockdown policy in Chinese polluted cities influenced the air pollution in the free troposphere. In this section, we attempted to quantify the net impacts of reduced anthropogenic emissions on BC concentrations at the receptor site. BC concentration is affected by both emission sources and meteorological conditions, such as temperature, relative humidity, wind speed and boundary layer height. Here, a random forest model (RF, TreeBagger in Matlab) was used to simulate the concentrations of BC. RF is an ensemble decision tree machine learning method which is a fusion of statistics, data science and computing. RF model can be used to isolate the contribution of meteorology and emission to air pollutant (Grange et al., 2018; Grange and Carslaw, 2019). In this model, meteorological conditions and CO concentrations (both BC and CO from similar sources) served as input. The number of variables used to grow a tree was set to be 1, the minimum node-size or depth was 5 and the number of trees was set to be 200. The correlation coefficient (R²) of measured BC and simulated BC was 0.95 (p < 0.05) with an Root Mean Square Error (RMSE) = 0.05 μg m⁻³ (Fig. S2). Figure S4 shows the relative importance of meteorological conditions (temperature, relative humidity, wind speed and boundary layer height) and emission sources to BC at WLG during the sampling period. Our result showed that CO was a major factor affecting the BC concentrations at WLG, suggesting the emission sources played an important role in controlling the BC concentrations at this mountainous site. To quantify the contribution of reduced emissions to BC at WLG, the average values of temperature, relative humidity, wind speed and boundary layer height before the lockdown would serve as input in the RF model to predict the BC concentrations in the pre-, during and post-lockdown. This reflected that the meteorological conditions did not change during the different sampling period. In other words, the difference of predicted BC concentrations between in the pre- and during COVID-19 lockdown was the response of BC concentration to the reduced anthropogenic emissions. As illustrated in Fig. 6, the average BC concentration from emission sources (BCemission) during the lockdown period was 0.20 ± 0.12 μg m⁻³, which was 0.09 μg m⁻³ lower than that (0.28 ± 0.19 μg m⁻³) in pre-lockdown period. The difference of BCemission between the pre- and during the COVID-19 lockdown accounted for 86% of the total decreased BC concentrations. This reflected that the reduced anthropogenic emissions were a major factor to lower the BC concentrations at WLG during the lockdown period. After the lockdown, the BCemission concentration rose to 0.23 ± 0.13 μg m⁻³, reflecting the re-operation of human activities and increased the BC concentrations contributed by the anthropogenic emissions.

![Fig. 5. Diurnal cycles of BCBB and BCFF at WLG during the different sampling periods.](image)

![Fig. 6. The average concentrations of observed BC (BC), simulated BC (BC_sim) and BC by emissions (BCemission) at WLG before, during and after lockdown.](image)
3.5. The potential source region of BC and CO\(_2\) moving towards sampling site

Based on the air cluster and CWT analyses (Fig. 7), we found that WLG station was mainly influenced by western airflow without significant contributions of anthropogenic emissions since southern Xinjiang and western Qinghai City are sparse population areas. Still, the nomadic people in the west part of Qinghai mainly burn cow dung and biomass for heating and cooking. Although small parts of air parcels were from the eastern region of WLG, the air masses passed over some intensive emission regions, such as Lanzhou and Xining, probably picking up the more polluted air to the sampling site. Consequently, the higher contributions of BC and CO\(_2\) were found when the air parcels from the eastern areas. This was consistent with the previous studies which concluded that the air pollution at WLG was mainly from Lanzhou-Xining areas (S. Liu et al., 2021a).

Fig. 8 illustrates the CWT results of BC (BC\(_{\text{CWT}}\)) at WLG of eastern and western airflows before, during, and after the lockdown. When the air parcels were from the eastern region, the average BC\(_{\text{CWT}}\) was \(0.211 \pm 0.178 \ \mu g \ m^{-3}\) in pre-lockdown. However, the BC\(_{\text{CWT}}\) decreased by 80% during the lockdown period (0.028 \(\pm 0.051 \ \mu g \ m^{-3}\)). Compared to the pre-lockdown, CO\(_2\)\(_{\text{CWT}}\) also decreased by 52% during the lockdown period. This suggested that the reduced anthropogenic emissions via the COVID-19 lockdown policy in the eastern region did decrease the BC and CO\(_2\) concentrations at WLG. On the contrary, the less decreased BC and CO\(_2\) concentrations when westerly airflow prevailed at the receptor site. This might reflect that less BC and CO\(_2\) emissions by traffic and industries sources were contributed from the western region of WLG since there were no populated cities located in the western region of Qinghai province. Based on the CWT analysis, it was found that the BC and CO\(_2\) decreased during the lockdown period when the air masses from the eastern region of WLG (97% of the total decline in BC, and 108% of the total decline in CO\(_2\)). However, no substantial differences of BC and CO concentrations were found as the air parcels came from the eastern areas. This suggested that the COVID-19 lockdown in the eastern region of WLG did result in the decreasing BC and CO\(_2\) concentrations at this mountainous site.

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

BC has positive radiation forcing effects and the reduction of BC concentrations during the lockdown period would directly change solar energy absorbance in the atmosphere. In this work, we used a simplistic model to calculate the reduction of solar energy absorbance by BC (\(E_{\text{BC}}\),

![Fig. 7. Air clusters and CWT of BC (a, c, e), and CO\(_2\) (b, d, f) at WLG in pre-, during- and post-lockdown.](image-url)
BrC_{pri} (E_{BrC-pri}), and BrC_{sec} (E_{BrC-sec}) at the wave of 300–950 nm due to the lockdown (Bosch et al., 2014; Liu et al., 2019). During the entire sampling period, the average total solar energy absorbance (E_{Total}) by carbonaceous aerosols was 2.96 ± 2.37 W m^{-2}. BC was the dominant species, which contributed approximately 75% to E_{Total}, followed by BrC_{pri} (13%) and BrC_{sec} (12%). The ratio of E_{BrC}/E_{BC} was 34 ± 20%, which was higher than that in Nanjing (17 ± 0.7%(Y.-C. Lin et al., 2021)), indicating that the fraction of BrC to solar energy absorbance was much higher at WLG. This might be caused by the lack of large industrial plants around the WLG site and biomass burning for herders around the site. During the lockdown, the solar energy absorbance by BC was 1.50 ± 1.29 W m^{-2}, which decreased by 35–37% compared to those in the pre- (2.33 ± 2.08 W m^{-2}) and post-lockdown (2.41 ± 1.68 W m^{-2}). Similarly, the E_{BrC_{pri}} and E_{BrC_{sec}} during the lockdown period also decreased by 15% and 14% (Table 1). The declined solar energy absorbance by the three species decreased the E_{Total} by 30% compared to the pre-lockdown. This highlighted that the reduced anthropogenic carbon emissions during the lockdown not only altered the concentrations of carbonaceous aerosols, but also decreased their solar energy absorbance in the Qinghai Plateau free troposphere.

**4. Conclusions**

In this study, we used a 7-band Aethalometer to continuously measure the absorption by carbonaceous aerosols from 3rd January to March 31, 2020 at WLG station in the north-eastern Tibetan Plateau region of China. The study period consisted of 89 days, including 31 days of pre-lockdown, 17 days of lockdown and 41 days of post-lockdown. We also calculated the BC masses from the observed light absorption data sets. Our result revealed that the average BC concentration was 0.18 ± 0.16 μg m^{-3} during the lockdown period, which decreased by 35% (with 15% due to meteorological conditions and 85% due to a decrease in emissions) compared with those in the pre-lockdown and post-lockdown. BC_{BB} decreased by 18% and BC_{FF} by 47% (20% and 80% of total BC decrease). During the COVID-19 lockdown period, the PM_{2.5} mass at WLG decreased by 39%. The lockdown also resulted in decreases of the solar energy absorbance by BC (decreasing rate was 35%), primary BrC (15%) and secondary BrC (14%). According to CWT analysis, the potential source region of BC at WLG was in Xining - Lanzhou urban cities which were located in the eastern part of China. During the lockdown period, BC_{CWT} decreased by 80% (95% of total decrease of BC_{CWT} during lockdown) and CO_{2 CWT} decreased by 52% (108% of total decrease of CO_{2 CWT} during lockdown) in the east, but there was no...
significant change observed during the whole sampling period from the west. The long transportation from city region in the east of site may cause the WLG aerosol results to be overestimated by about 30%, but in the case of CO₂, may cause just about 1 ppm overestimation.

COVID-19 lockdown provided a good platform to explore the response of air pollution to the reduced anthropogenic emissions in urban cities. It also supplied the opportunity to influence the investigation of anthropogenic emissions on atmospheric chemistry in the free troposphere. WLG is one of the GAW/WMO baseline stations in the world and it supplied the worthy monitoring data to explore the changes of the characteristics of real background atmosphere, which was isolated from the influence of the anthropogenic emissions from surface ground, and the interactions between air pollution and climate. However, we found an impact by anthropogenic emissions from the surrounding downwind populated cities, suggesting the regional background atmosphere was also influenced by the emissions from surface ground. Thus, how to screen out the influence of anthropogenic emissions on air pollution will be an important issue for WLG, as well as the feedbacks of climate change to the atmospheric chemistry in the free troposphere over the Qinghai Plateau region.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.envres.2022.112984.

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