Characteristics of airborne black carbon-containing particles during the 2021 summer COVID-19 lockdown in a typical Yangtze River Delta city, China

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Abstract

Black carbon-containing particles (BCc) are ubiquitous in ambient air, significantly contributing to particulate matter (PM) pollution. The unexpected outbreak of the COVID-19 pandemic in the summer of 2021 prompted a localized and prolonged lockdown in Yangzhou City, situated in the Yangtze River Delta, China. This lockdown led to significantly altering in local anthropogenic emissions, while neighboring cities continued regular operations, providing a unique opportunity for the investigation of BCc characteristics influenced by varying emission conditions. Single particle aerosol mass spectrometer (SPA-MS) analysis revealed a notable decrease in the proportion of freshly emitted BCc during the lockdown period (LD). However, $PM_{2.5}$ concentrations remained relatively unchanged, with an observed increase in the proportion of aged BCc during LD compared to the period before the lockdown (BLD). The study also underscored the significant role of regional transport in $PM_{2.5}$ pollution during the campaign. Moreover, reactive trace gases (e.g., NO_x, SO_2, and VOCs) could form thick coatings on pre-existing particles likely via enhanced heterogeneous hydrolysis under high relative humidity (RH) as well, resulting in significant BCc particle growth (~600 nm), as well as $PM_{2.5}$, during LD. Our study highlights that short-term, strict local emission controls may not effectively reduce PM pollution due to the complex production and transmission characteristics of BCc and the non-linear responses of $PM_{2.5}$ to its precursors. Achieving further effective $PM_{2.5}$ reduction mandates a focus on nuanced control of BCc and necessitates a comprehensive and extensive approach with a regionally coordinated and balanced control strategy through joint regulation.

1. Introduction

China has implemented long-term clean air measures to cut down anthropogenic emissions and improve air quality (Ge et al., 2020), resulting in a nationwide reduction of average fine particulate matter ($PM_{2.5}$, aerodynamic diameter ≤ 2.5 μm) level from 50 μg m^{-3} in 2015 to 30 μg m^{-3} in 2020 (Zhou et al., 2022). However, this $PM_{2.5}$ concentration remains significantly higher than the new World Health Organization (WHO) guideline value of 5 μg m^{-3} (*WHO Global Air Quality Guidelines*, 2021). Black carbon (BC) is a ubiquitous component of aerosols, typically constituting a small proportion (5~10%) of $PM_{2.5}$ in the atmosphere (Chen et al., 2020). However, freshly emitted BC evolves into BC-containing particles (BCc) by undergoing atmospheric aging, contributing to a rise in the total mass of $PM_{2.5}$ through processes of coating or embedding by other materials (Bond and Bergstrom, 2006; Peng et al., 2016). The number and mass fraction of BCc can excess 60% and 50% of $PM_{2.5}$, respectively, emphasizing the significant role of BC in elevating the mass concentration of particulate matter (PM) (Sun et al., 2022; Xie et al., 2020; Chen et al., 2020).

The atmospheric aging of BCc involves intricate chemical and physical transformations that influence their mixing state, morphology, hygroscopicity, and optical properties, all of which have profound implications for climate and human health (Bond et al., 2013; Ramanathan et al., 2008). For example, freshly emitted BC particles are initially hydrophobic but possess a porous surface structure that facilitates the internal or external mixing with co-emitted primary organic/inorganic and secondary materials that are associated with BC (Cheng et al., 2012; Li et al., 2020). On the other hand, BCc undergoes continually aging processes, including the condensation of low-volatility vapors (Li et al., 2022), coagulation with preexisting aerosols (Kondo et al., 2011), and heterogeneous oxidation with gaseous pollutants (Zhang et al., 2024). This alteration may affect the coating thickness, morphology, size distribution, and hygroscopicity of BCc, thereby impacting their climate forcing as well as atmospheric lifetime (Luo et al., 2022; Taylor et al., 2014). High loading of atmospheric BCc could also depress the development of the planetary boundary layer and exacerbate PM pollution episodes (Huang et al., 2018). BCc characteristics are influenced by various combustion sources and emission conditions, including local industrial burning, vehicle exhausts, residential coal burning, and biomass burning (Li et al., 2020; Sedlacek et al., 2022; Zhang et al., 2018), as well as long-range transport from other regions (Adachi et al., 2014; Zhang et al., 2021). Those diverse conditions complicate the development of parameterizations of BCc properties, the insufficient understanding of complex emission sources, aging processes, and physical properties of BCc, hampering the effectiveness of air quality remediation (Cappa et al., 2019; Kahnert, 2010; Sun et al., 2021).

Studies on the effects of large-scale and short-term stringent emission control events on air quality in China have been widely deployed, e.g., the 2008 Beijing Olympic Games

(Wang et al., 2010; Zhou et al., 2010), the 2015 Asia-Pacific Economic Cooperation (APEC) (Zhu et al., 2015), the 2014 Nanjing Youth Olympic Games (Wang et al., 2022) and the national COVID-19 lockdown in 2020 winter (Huang et al., 2021; Le et al., 2020; L. Li et al., 2020; Wang et al., 2020). Previous studies extensively investigated air pollutant variations during the COVID-19 lockdown in the winter of 2020 across different regions of the world. Stringent restrictions on industrial and vehicular activities have resulted in significant reductions in gaseous pollutants and particulate matter, not only in megacities (Chen et al., 2020; Jeong et al., 2022; Sun et al., 2020) but also in middle-sized cities (Clemente et al., 2022; Wang et al., 2021; Xu et al., 2020) and rural areas (Cui et al., 2021, 2020; Jain et al., 2021). Compared to the decreasing trends observed in most cities worldwide, the level of $PM_{2.5}$ in Shanghai (Chang et al., 2020), Hohhot (Zhou et al., 2022), and the Northeast of China Plain (Nie et al., 2021) increased unexpectedly. These observations reveal the complex aerosol chemistry of $PM_{2.5}$ comprising primary and secondary components. The reduction of primary pollutants during lockdown resulted in a shift towards a higher proportion of secondary aerosols, including inorganic and organic species, exhibiting a non-linear response to emission changes (Zhang et al., 2021). Furthermore, some studies suggested that the increase in secondary aerosols during lockdown is due to the enhanced atmospheric oxidative capacity resulting from the rise in ozone levels (Wang et al., 2021), unfavorable meteorological conditions (Chien et al., 2022; Sulaymon et al., 2021a), changes of local and regional emission sources (Feng et al., 2022). However, most previous studies focused on lockdown events during the cold seasons, and studies on summer lockdown events in China were very limited.

Yangzhou is located in the central region of the Yangtze River Delta (YRD), at the junction of the Yangtze River and, the Beijing-Hangzhou Grand Canal, which serves as a prominent economic city, industrial-intensive area, and highly active inland shipping node in East China. Due to the complex emissions and feedback with the East Asian monsoons (Ding et al., 2019), this region is susceptible to anthropogenic aerosols, especially BCc originating from chemical, steelmaking, coal-fired, petrochemical enterprises, and transportation, etc. Extensive studies have investigated the responses of atmospheric pollutants to emission changes during the COVID-19 lockdown measures in the YRD (Chen et al., 2021; Li et al., 2020; Qin et al., 2021; Zhang et al., 2022). However, the key chemical and physical processes specifically responsible for the BCc in this region are still unclear. During the summer of 2021, Yangzhou experienced a resurgence of COVID-19 with over 500 confirmed cases. In response, stringent public health measures were imposed from July 29th to September 10th, including the closure of public transport, and suspension of non-essential industrial plants, restaurants, shopping malls, and entertainment clubs. People were also mandated to quarantine at home. Consequently, Yangzhou experienced a significant decline in transportation and industrial energy consumption, dropping by nearly 46% and 25%, respectively, compared to the same period in 2020 (www.yangzhou.gov.cn),

implying a substantial reduction in human activity and primary emissions. Unlike the nationwide COVID-19 lockdown in China during the cold season of 2020 (Le et al., 2020; Sulaymon et al., 2021b), the summer lockdown in Yangzhou was more localized but protracted, significantly altering local anthropogenic emissions while neighboring cities maintained regular operations, which provides a unique opportunity to explore and compare the diverse mixing states and, the aging process of BCc in different anthropogenic emission conditions in summer. Here we report the chemical compositions and aging characteristics of airborne BCc in YRD. Our investigation involved a combination of ground measurements, spaceborne observations, and mass spectrometric analysis conducted during the COVID-19 lockdown in the summer of 2021 in Yangzhou. Additionally, we employed potential source contribution function (PSCF) analysis to investigate the air pollution patterns in the YRD.

2. Methods

2.1 Sampling site and instruments

The in-situ online measurements were conducted at a rooftop laboratory 20 m above ground located in a national air quality monitoring station, Yangzhou Environmental Monitoring Center (32.41ºN, 119.40ºE), Yangzhou, China (**Figure 1**). This sampling site is a typical urban site surrounded by residential areas, arterial roads, parks, restaurants, and shopping centers. In this study, the measurement period was divided into three phases: the before-lockdown period (BLD: 30 June to 28 July 2021), the lockdown period (LD: 29 July to 9 September 2021), and the after-lockdown period (ALD: 10 September to 7 October 2021) (**Figure 2**).

A single-particle aerosol mass spectrometer (SPA-MS, Hexin Analytical Instrument Co., Ltd., China) was deployed during the field campaign to obtain the chemical composition, size distribution, and mixing state of individual $PM_{2.5}$ particles. A cyclone with a 2.5 μm cutpoint (Model URG-2000-30ED) and a Nafion dryer is equipped in front of the sampling inlet. Individual particles are introduced into the SPA-MS through a critical orifice at a flow rate of 3 L min^{-1}. The vacuum aerodynamic diameters (D_{va}) are determined using the velocities derived from two continuous laser beams (diode Nd: YAG, 532 nm) spaced 6 cm apart. Subsequently, these particles are desorbed and ionized by a downstream pulsed laser (266 nm), and ion fragments are generated and measured by a Z-shaped bipolar time-of-flight mass spectrometer. A more detailed description of SPA-MS can be found in previous studies (Li et al., 2011).

$PM_{2.5}$ mass concentration was measured by a particulate matter monitor (XHPM2000E, Xianhe, China). Nitrogen oxides (NO_x = NO + NO_2), SO_2, and ozone (O_3) concentrations were detected with a set of Thermo Fisher Scientific instruments (Models 42i, 43i, and 49i). The concentrations of 103 volatile organic compounds (VOCs) in ambient air, comprising 57 ozone precursors (PAMS), 12 aldehydes and ketones, and 34 toxic organics (TO15), were continuously monitored at hourly intervals

using an online device (TH-300B, Tianhong, China). Meteorological parameters, including ambient temperature (T), relative humidity (RH), wind direction (WD), and wind speed (WS) were observed synchronously using an automatic weather instrument (WXT530, Vaisala, Finland). Precipitation (PCP) data was obtained from the Yangzhou Meteorological Bureau. All online data presented in this paper were hourly averaged at local time (Beijing time, UTC+8).

2.2 Data analysis

2.2.1 Satellite Product

In this study, we utilized the Copernicus Atmosphere Monitoring Service (CAMS) Global Near-Real-Time dataset (available at https://developers.google.com/earth-engine/datasets/catalog/ECMWF_CAMS_NRT), acquired from the European Centre for Medium-Range Weather Forecasts (ECMWF), to analyze the distribution of total surface column concentrations of NO_2, SO_2 and surface $PM_{2.5}$ mass concentration. CAMS offers the capacity to continuously monitor the composition of the Earth's atmosphere at global and regional scales since 2016, with a spatial resolution of 44528 meters (Benedetti et al., 2009; Morcrette et al., 2009). The details of the bands of the dataset used in this study are shown in Table S2. We calculated and plotted the averaged 2-dimensional data of ECMWF/CAMS/NRT NO_2, SO_2, and $PM_{2.5}$ during BLD and LD over the region of interest (17.93~54.74 ºN, 71.21~142.23 ºE) using Google Earth Engine (Gorelick et al., 2017). The integration of remote sensing measurements has provided a more comprehensive understanding of the sources and distributions of particle matter and gaseous pollutants facilitating the evaluation of the impact of human activities on air quality.

2.2.2 Geographic Source Analysis

The potential source contribution function (PSCF) analysis, based on the Hybrid Single-Particle Lagrangian Integrated Trajectory (HYSPLIT) model, can be employed to identify regional sources of air pollutants. Before conducting the PSCF analysis, 36 hours of air mass backward trajectories with one-hour resolution at 500 m above ground level were calculated using the wind data from the Global Data Assimilation System (GDAS) provided by the National Oceanic and Atmospheric Administration (NOAA) (Wang et al., 2009). An open-source software MeteoInfo (Wang, 2014) was utilized for the PSCF analysis. The whole study area (110.1~133.4 ºE and 21.3~39.9 ºN) covered by the trajectories was divided into thousands of cells with a spatial resolution of 0.1° × 0.1°. The PSCF was simulated according to the following equation:

$$PSCF_{ij} = \frac{m_{ij}}{n_{ij}} \quad (1)$$

where $PSCF_{ij}$ is the conditional probability that the grid cell (i, j) was a source of the species found in high concentration (Hopke et al., 1993); n_{ij} is the number of all trajectories passing through this grid cell, and m_{ij} is the number of trajectories. In this study, the pollution criterion values for different BCc particle types were set as the 75th

percentile of hourly average number fractions, respectively. To further improve the accuracy of the PSCF analysis and minimize analytical uncertainties, the Weighted PSCF (WPSCF) functions as shown in Equation (2~3) were applied (Polissar et al., 1999). The weight (W_{ij}) for each grid cell was determined based on the number of trajectory endpoints (n_{ij}) as follows:

$$WPSCF_{ij} = W_{ij} \times PSCF_{ij} \quad (2)$$

$$W_{ij} = \begin{cases} 1.00 & n_{ij} > 3n_{ave} \\ 0.70 & 1.5n_{ave} < n_{ij} \leq 3n_{ave} \\ 0.40 & n_{ave} < n_{ij} \leq 1.5n_{ave} \\ 0.17 & n_{ij} \leq n_{ave} \end{cases} \quad (3)$$

Here, n_{ave} is the average number of trajectory endpoints of each grid.

2.2.3 SPA-MS Data Analysis

In total, 1649574 particles were analyzed during the entire observation period. The size and chemical composition of single particles were analyzed using the Computational Continuation Core (COCO V1.4) toolkit in MATLAB 2022 (The MathWorks, Inc.). Our focus was on BCc, which was identified based on the relative peak area (RPA) of carbon ion clusters ($C_n^{\pm}$, n = 1, 2, 3, …), with a threshold of 0.05 (Zhang et al., 2021). An adaptive resonance theory-based neural network algorithm (ART-2a) was applied to classify the measured individual particles based on the presence and intensity of ion peaks, with a vigilance factor of 0.75, a learning rate of 0.05, and 20 iterations (Song et al., 1999).

3. Results and discussion

3.1 Field observations

Figure 2 presents the temporal variations of meteorological parameters, $PM_{2.5}$, NO_x, and SO_2 concentrations. Notably, significantly reductions in $PM_{2.5}$, NO_x, and SO_2 were observed at the end of BLD due to a high precipitation event, with a peak hourly precipitation reaching 37 mm, and the data collected during this event were excluded from the analysis. During BLD, the mean temperature (T) was 28±3 ℃, the total precipitation was 221 mm, with an average relative humidity (RH) of 83±12%. The prevailing winds originated from the south and southeast, with a mean wind speed (WS) of 3.3±1.2 m s^{-1}. In comparison, LD shows a decline in temperature to 27±2 ℃ and WS to 2.3±1.0 m s^{-1}, but an increase in RH to 87±11% and a reduction in total precipitation to 86 mm. **Figure S2b and c** present uniform distributions of RH and boundary-layer height (BLH) across the YRD during LD. These regional meteorological conditions and the effective removal of the pollutants accumulated at the end of BLD facilitated the investigation of BCc regional transport in YRD. During ALD, the temperature declined further to 25±3 ℃, WS increased to 3.3±1.5 m s^{-1}, and total precipitation dropped to 27 mm with a lower RH of 77±14%.

During LD, strict measures resulted in notably lower surface concentrations of $PM_{2.5}$ (20.3 μg m^{-3}), NO_x (16.8 μg m^{-3}) and TVOC (55.9 μg m^{-3}) compared to BLD and ALD. Conversely, the surface O_3 concentration showed an increase of 18.4 μg m^{-3} (28%) during LD relative to BLD. The reduction of fresh NO emission alleviates O_3 titration (Steinfeld, 1998) could be an explanation. Analysis from **Figure S3** indicates that the O_3 level is higher than those of neighboring cities in the YRD, suggesting higher local atmospheric oxidation capacity during LD. However, the average concentrations of $PM_{2.5}$ (20.6 vs. 20.3 μg m^{-3}), SO_2 (9.1 vs. 9.2 μg m^{-3}) and CO (0.61 vs. 0.62 mg m^{-3}) were comparable during both BLD and LD (**Figure 3**).

After LD, social activities gradually resumed in Yangzhou City, leading to an apparent increase in all observed pollutants during the ALD period. For instance, there were relative increases of 66% for NO_x, 19% for SO_2, 36% for TVOC, 14% for O_3, 32% for $PM_{2.5}$, and 16% for CO from LD to ALD, respectively (**Figure 3**). ~~Given that both BC and CO are byproducts of incomplete combustion of carbon-containing fuels (Wang et al., 2015), and the high correlation between BC and CO (Zhou et al., 2009), it is plausible to infer that the primary emission source of BC during LD differed from those during ALD. This change likely reflects the shift in combustion practices and fuel usage patterns as economic activities restarted during ALD.~~

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Satellite-retrieved $PM_{2.5}$, NO_2, and SO_2 data over the entire region of eastern China were also investigated, and results show that these pollutants were predominantly concentrated in Shanghai and its neighboring cities, including Yangzhou, during both BLD and LD (**Figure S4**). **Figure 4** presents regional fractional changes of mean $PM_{2.5}$, NO_2, and SO_2 concentrations from the BLD to LD periods in YRD, all showing an increase of 29%, 6%, and 14%, respectively. In comparison, Yangzhou city experienced lower increases in these air pollutants, with slight changes of 6%, -18%, and -4% for $PM_{2.5}$, NO_2, and SO_2, respectively. The implication is that, even though local primary emissions, such as NO_2, and SO_2, were reduced substantially during LD, they still could be affected by regional transport. Furthermore, as depicted in **Figure S3**, the concentrations of NO_2 in major cities of the YRD were more than twice higher than in Yangzhou during LD, confirming a relatively lower local primary emissions due to the stringent lockdown. However, the higher level of SO_2 in Yangzhou during LD may be attributed to the nearby power stations along the Yangtze River, which were not impacted by the lockdown measures.

3.2 Chemical composition and size distribution of individual BCc

Based on the SPA-MS analysis, a total of 1068362 BCc was collected during the whole study period. The BCc accounted for 59%, 69%, and 57% of the total number of measured particles in the BLD, LD, and ALD periods, respectively. **Figure 5** shows the normalized average mass spectra of BCc during three periods. Ion height in each spectrum reflects the number fraction of the detected BCc with the corresponding ion

to the total BCc, while colors represent peak area ranges of detected ions. BCc in BLD, LD, and ALD shown similar mass spectra at *m/z* < 100, with common peaks including carbon ion clusters ($C_n^{\pm}$, n = 1~7), m/z 27$[C_2H_3]^+$, 37$[C_3H]^+$, 43$[C_2H_3O]^+$, 51$[C_4H_3]^+$, 63$[C_5H_3]^+$, 46$[NO_2]^-$, 62$[NO_3]^-$, and 97$[HSO_4]^-$. However, the abundance of large *m/z* carbon ions ($C_n^{\pm}$, n > 7) in both BLD and ALD periods was 1.5 times higher than that in the LD. Previous studies have indicated that high-mass carbon ions may be linked to traffic emissions, particularly those from diesel trucks(Xie et al., 2020; Liu et al., 2019), and the observed reduction in such ions during LD suggests a decrease in local vehicle emissions. This trend is also consistent with the changes observed in aromatic compounds, e.g. m/z 119$[C_9H_{11}]^+$.

Further, BCc was classified into 12 types based on the differences in chemical features and temporal variations, as shown in **Table S1**. Fresh BC particles (BC-fresh) are those freshly emitted without undergoing significant atmospheric processing (Ding et al., 2021). Five types of BC-fresh particles were identified according to their ion markers: (i) BC-pure is dominated by carbon clusters ($C_n^{\pm}$) with minor ion signals of inorganic species, such as m/z 46$[NO_2]^-$ and m/z 97$[HSO_4]^-$ from nitrate and sulfate, respectively (Xie et al., 2020); (ii) BCc from biomass burning (BB) are characterized by ion signals at *m/z* 39$[K]^+$, 45$[CHO_2]^-$, 59$[C_2H_3O_2]^-$, and 73$[C_3H_5O_2]^-$, with a relative peak area (RPA) more than 0.5 (Silva et al., 1999); (iii) coal combustion BCc (CC) typically include small carbon clusters ($C_n^{\pm}$, n = 1~4), metal elements (e.g., m/z 7$[Li]^+$, 23$[Na]^+$, 27$[Al]^+$, 56$[Fe]^+$, 63$[Cu]^+$ and 206/207/208$[Pb]^+$), and organic carbon (38$[C_3H_2]^+$, 43$[C_2H_3O]^+$) peaks in the positive mass spectrum, while the strong signals of secondary inorganic species (46$[NO_2]^-$, 43$[AlO]^-$, 62$[NO_3]^-$, 80$[SO_3]^-$, 97$[HSO_4]^-$) in the negative ion mode suggest that CC particles were long-distance transported or more processed (Zhang et al., 2022; Zhang et al., 2009); (iv) particles from vehicle emission (VE) are characterized by the presence of ion signals at *m/z* 40$[Ca]^+$, 51$[V]^{+\cdot}$ 55$[Mn]^+$, 67$[VO]^+$, 46$[NO_2]^-$, 62$[NO_3]^-$, and 79$[PO_3]^-$, as well as high loadings of organic carbon (41$[C_3H_5]^+$, 43$[C_2H_3O]^+$) and carbon clusters ($C_n^{\pm}$, n = 1~4) ion peaks (Yang et al., 2017); (v) BCc that are internally mixed with more than one type (BB, CC, and VE) are categorized as Mix type (Sun et al., 2022).

Aged BC particles, denote as BC-aged, undergo a series of chemical reactions and physical transformations. These processes typically lead to changes in their morphology, hygroscopicity, and optical properties as they are coated with other materials (He et al., 2015). Six types of BCc are classified as BC-aged and are further grouped into BCOC and BC-SNA, depending on whether they contain mainly organic carbon (OC) or sulfate/nitrate/ammonium (SNA). First, BCOC types indicate BC-aged particles that are internally mixed with OC. These particles are characterized by the presence of carbon clusters ($C_n^{\pm}$) and $C_nH_m^+$ ions (n = 1~6, m = 1~3) in positive mass spectra (Xie et al., 2020). On the other hand, BC-aged particles that do not mix with OC are named BC-SNA indicating the mix with secondary inorganic species. Additionally, BCOC

particles with negative mass spectra dominated by nitrate ions (46[NO_2]$^-$ and 62[NO_3]$^-$) or sulfate ions (97[HSO_4]$^-$) are referred to as BCOC-N or BCOC-S, respectively; otherwise, BCOC particles showing similar peak areas of nitrate and sulfate are named BCOC-SN. The BC-SNA particles are further categorized as BC-N, BC-S, and BC-SN based on similar principles. Note the remaining particles that cannot be classified into either BC-fresh or BC-aged ones are denoted as BC-other. More details of BCc particle types are shown in **Table S1** and **Figure S1** in the Supplement.

During BLD, the average number fraction of BC-fresh particles was 36% with sizes mainly concentrated at 500 nm, similar to the mode size of BC-aged particles was 520 nm (**Figures 6**). The predominant BCc types during BLD were BCOC-S and BC-S (24% and 12% by number), likely because sulfate was removed less efficiently than organic matter (OM) and NO_3 by heavy precipitation, especially during the warm seasons (Isokääntäet al., 2022). As shown in **Figures 6c and d**, the peak size of BC-SNA was larger than that of BCOC in all periods, indicating that organics coated BCc generally had a relatively thin coating compared to those coated by secondary inorganic species, which is consistent with previous studies (Sun et al., 2016; Wang et al., 2019).

During the transition from BLD to LD, heavy and continuous precipitation occurred from July 25th to July 28th (the eve of lockdown), resulting in the removal of a majority of the pollutants ($PM_{2.5}$: 4 μg m^{-3}, O_3: 35 μg m^{-3}, NO_x: 8 μg m^{-3}). Following this environmental clearance, strict lockdown measures were implemented, resulting in a drastic reduction in primary emissions. As a result, the number fraction of BC-fresh particles significantly decreased from 37% to 28% and that of VE-type particles dropped from 12% to 3% (by number). Expectedly, with the decrease in NO_x, an obvious enhancement of O_3 was observed during LD (**Figure 3**). According to previous studies (Huang et al., 2021; Laughner et al., 2021), large reduction of NO_x may promote the formation of O_3 under a VOC-limited regime and enhance the oxidation capacity of the local atmosphere, which may promote the number fraction of BC-aged particles increased from 64% in the BLD to 72% in LD (**Figure 7a**), indicating the lockdown could accelerate aging of BCc through complicated chemical reactions and/or physical coagulation. Additionally, the most abundant type of BCc changed from BCOC-S (24% by number) in the BLD to BC-N (25%) in LD (**Figure 7a**), suggesting different BCc formation pathways. Despite the abrupt reductions of NO_x (-39%) due to the city lockdown, it is important to note that the concentration of $PM_{2.5}$ only slightly decreased during LD (-1%), highlighting the non-linear relationship between primary emissions and $PM_{2.5}$ levels.

During ALD ($PM_{2.5}$: 26.7 μg m^{-3}, NO_x: 27.9 μg m^{-3}, TVOC: 76.0 μg m^{-3}), the number fraction of BC-fresh particles rose from 28% (LD) to 31% (ALD), while the fraction of VE particles also increased from 3% (LD) to 12% (ALD) (**Figure 7a**), coinciding with a 16% rise in CO concentration. This suggests a shift in combustion practices and fuel

usage patterns as economic activities resumed. The increased CO levels, a known marker for combustion-related emissions, align with the resurgence of vehicle emissions and other activities that emit BC and CO concurrently (Wang et al., 2015, Zhou et al., 2009). Notably, the size distributions of BC-fresh and BC-aged particles presented relatively small peaks at 690 nm and 820 nm during ALD, in addition to the prominent peaks at 490 nm and 500 nm, which were different from those in the BLD and LD periods. These small peaks were relatively close to the dominant sizes of BC-fresh and BC-aged particles during LD (**Figure 6**). This result suggests that a substantial number of BCc with small sizes (around 500 nm) after the lockdown was lifted in Yangzhou, owing to the sudden enhancement of primary emissions; on the other hand, particles with large diameters (>690 nm) may have formed due to the participation of more trace reactive gases (e.g., NO_x, SO_2, and VOCs) in continuous aging reactions, resulting in thicker coatings on the surface of pre-existing particles and therefore a more clear separation of two-mode sizes during the ALD period than during the other two periods. This hypothesis was also supported by the increased number fraction of BCOC-SN during the ALD period (**Figure 7a**). Similar findings have been reported in the North China Plain (NCP) and the YRD during cold seasons, where thicker coatings on secondary aerosols were also observed under lower RH (<70%) (Zhang et al., 2021). This might be due to that particles with more organics and nitrate can result in earlier deliquescence and provide aqueous surfaces that facilitate the heterogeneous formation of secondary species under relatively low RH (Zhang et al., 2021). Among the three periods, the difference between the mode sizes of BC-aged and BC-fresh particles was the smallest (10 nm) during the ALD period (**Figure 6a and b**). This size reduction can be attributed to the increased BCOC and hydrophobic primary particles after lockdown (Figure 7). Because the internally mixed BCOC and hydrophobic primary particles may constrain further growth of secondary BC-SNA particles (Liu et al., 2016; Zhang et al., 2018), thereby leading to smaller-sized BC-aged particles. Moreover, the differences in BCc mode sizes between ALD and BLD periods also reveal an interesting fact that the lockdown effect may not only affect air quality during lockdown but also can influence the air quality even after lockdown, as the resumed emissions after lockdown may be subjected to different chemistry from that before lockdown.

Throughout the entire observation, the changes in the number fraction of BC-SNA exhibited consistency with the variations in RH (**Figure 7b**), indicating that BC tends to mix with ammonium sulfate and ammonium nitrate under high RH conditions. Meanwhile, the number fraction of BCOC shows similar patterns as TVOC, suggesting that high TVOC levels may facilitate the coating of organics on BC cores under low RH condition. **Figure 8** displays the number fraction of BCc species as a function of $PM_{2.5}$. Overall, as $PM_{2.5}$ levels increased, the number fraction of BC-aged particles also increased, while the proportion of BC-fresh particles decreased during BLD and LD, indicating a clear transition from BC-fresh particles to more aged ones, in line with the average size distribution during ALD has a small peak at 900 nm. Specifically, the

increase in $PM_{2.5}$ was driven by BCOC-S during BLD (**Figure 8a**), whereas BC-N played a vital role in the $PM_{2.5}$ increase during LD (**Figure 8b**). Interestingly, the concentration of NO_x, the primary precursor of BC-N, decreased by 31% and 41% during LD compared to BLD and ALD, respectively (**Figure 3)**, indicating a strong non-linear response of nitrate in BCc to NO_x, likely due to much faster conversion of NO_x to nitrate upon enhanced atmospheric oxidation capacity; additionally, the high proportion of BC-N during LD might be attributed to regional transport, similar to that in Shanghai during 2020 winter lockdown (Chang et al., 2020).

3.3 Chemical aging of BCc

As shown in **Figure 5**, in the average positive mass spectra of total BCc, the peak areas of C_n^+, OM, and metals contributed to more than 95% of the total, while nitrate and sulfate peak areas accounted for more than 90% of the negative mass spectral signal. To better elucidate the aging processes of BCc during different lockdown periods, we summed the carbon clusters $C_n^{\pm}$ (n = 1~5, accounting for more than 99% of C_n) peak areas to represent BC, and the total peak area of sulfate, nitrate, and ammonium (SNA) to represent the second inorganic components coated on BC. Additionally, we defined the sum of positive peak areas, excluding C_n^+ and metals, as OC to represent the OM coated on BC. These peak areas encompassed almost all the coating materials, except for metals, of BCc. The changes in the mixing state and morphology of BCc can provide insights into their aging characteristics, as reported previously (Kandler et al., 2018; Moffet et al., 2013). In this study, we use OC/C_n and SNA/C_n ratios to describe different types of chemical components coated on BC-fresh, and we use the ratio of the mode size of BC-aged (D_{aged}) to that of contemporaneous BC-fresh (D_{fresh}) to represent the aging degree of BCc.

Figure 9 illustrates the diurnal variations of the OC/C_n and SNA/C_n ratios along with the size distribution of BCc during different periods. We observed that both OC/C_n and SNA/C_n increased during nighttime and decreased during daytime. These variations showed the prominent enhancements of nocturnal OM and SNA, which could be attributed to the accelerated gas-to-particle partitioning and nocturnal secondary formation of organic/inorganic components under high relative humidity (RH > 85%) and relatively stagnant air mass (WS < 3 m s^{-1}) (**Figure S5**). It is worth noting that from BLD to LD and ALD, the intensity of diurnal variations of OC/C_n and SNA/C_n increased obviously. This discrepancy can be attributed to several reasons. (i) During BLD, the frequent precipitations effectively scavenged the particles (Isokääntä et al., 2022); (ii) In contrast, stronger solar radiation and higher O_3 concentration during LD promoted photochemical formations of OC and SNA; (iii) After lockdown, more precursors due to increased local emissions may lead to more production of secondary components than that during BLD as explained earlier. These results indicate that the aging process and mixing state of BCc depend strongly on meteorological conditions as well as emission sources in urban cities.

As shown in **Figure 9**, BCc with ~400 nm D_{va} exhibited significant diurnal fluctuations in the OC/C_n and SNA/C_n ratios, during LD. There is a noticeable increase in the proportion of BC-SNA particles during nighttime when RH is relatively high. These observations suggest that nighttime heterogeneous hydrolysis may be considered a key mechanism responsible for the formation of BCOC and BC-SNA particles. According to Jacobson (2002), coagulation can be significant between particles with sizes <100nm and >1μm but insignificant for particles of >300nm, when the total particle number concentration is higher than 10^4 cm^{-3}. During LD, the OC/C_n and SNA/C_n ratios of BCc with ~400 nm D_{va} exhibited pronounced diurnal variations (**Figure 9**) and the number fraction of BC-SNA increased obviously. Despite the difference between D_{va} and physical diameter, such results imply that chemical reactions should be considered as the major pathway for BCOC and BC-SNA particles of ~400 nm D_{va}, while the large-sized BC-aged particles (>1 μm) may be partially from physical coagulation. The more significant diurnal fluctuations in the OC/Cn and SNA/Cn ratios of BCc particles during the ALD period, compared to the LD period, can be attributed to increased primary emissions from resumed society activities, more complex atmospheric chemistry involving reactive gases, and the reinstatement of typical diurnal emission patterns, with higher nighttime RH further enhancing secondary aerosol formation.

According to Surdu et al. (2023), condensation involves the direct deposition of gas-phase molecules onto the surface of particles, driven by the difference between the condensable gases concentration (C_g) and its equilibrium particle-phase concentration (C_{eq}), which is negatively affected by RH. In our study, the average RH was relatively high during all three periods (>75%), but the condensable vapor concentration decreased during the lockdown period due to strict lockdown measures, making the difference between C_g and C_{eq} smaller during LD compared to the other two periods. Additionally, we observed a larger mode peak (600 nm, Dva) and higher Daged/Dfresh ratios (1.11) compared to BLD (510 nm, 1.03) and ALD (500 nm, 1.02) (Figure 6). Therefore, we conclude that condensation was likely inhibited during the LD period. Instead, the conditions likely favored aqueous-phase and heterogeneous reactions, which played a more important role in the evident growth of BCc particles, converting partially coated particles into fully thickly coated BCc during the LD period.

~~Additionally, the larger mode peak (600 nm, D_{va}) and higher D_{aged}/D_{fresh} ratios (1.11) were observed compared to those of BLD (510 nm, 1.03) and ALD (500 nm, 1.02) (**Figure 6**). Due to the decreased concentrations of the trace reactive gases (e.g., SO_2, NO_x, and VOCs) gases because of the strict lockdown measures and the higher relative humidity (RH > 75%) conditions, which depressed condensing process and facilitate the aqueous or heterogeneous reactions. might play a more important role leading to evident growth in the size of BCc. This aqueous or heterogeneous process during LD likely converted partially coated particles to fully thickly coated BCc as well.~~

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3.4 Source apportionment of BCc during lockdown

In addition to local emissions, regional transport plays a significant role in influencing pollutant levels. The emergent lockdown in Yangzhou led to strict limitation on local emissions, while surrounding cities were still running as usual. This is supported by **Figure S6**, which illustrates the $PM_{2.5}$ concentrations in Yangzhou and the other five surrounding YRD cities (e.g., Nanjing, Zhenjiang, Changzhou, Taizhou, and Chuzhou) during the campaign. High correlations between $PM_{2.5}$ concentrations in Yangzhou and the other five cities were observed across all different periods (**Figure S6**). These findings underscore the importance of the regional transport in $PM_{2.5}$ pollution during the campaign, providing a unique opportunity to investigate the transmission and source characteristics of BCc in YRD during summer. Herein, PSCF analysis was applied to qualitatively simulate the source probability distributions of the specific BCc particle types (BC-fresh, BC-aged, BCOC, and BC-SNA) during LD.

As shown in **Figure 10**, the hotspots of potential sources for the four particle types exhibited strong agreements with each other and primarily concentrated in the southeast of Yangzhou, especially along the coast of the Yangtze River, with the WPSCF greater than 0.6. These hotspot areas also encompassed chemical enterprises, power plants, petrochemical industrial parks, and the Yangtze River in the YRD. This evidence suggests that the region of southeast Yangzhou and lower reaches of the Yangtze River are major source areas for the regionally transported BCc in Yangzhou during lockdown. Additionally, Luo et al. (2023) reported that regional transport of pollutants can occur near the surface from upwind areas when the wind speed (WS) exceeds 2 m s^{-1}. **Figure S5b** shows that the mean daytime WS was 3 m s^{-1}, indicating that both BC-fresh and BC-aged particles, along with trace gases (e.g., SO_2, NO_x, and VOCs), originating from the hotspot areas, could be transported effectively to Yangzhou. Additionally, the average size of BCc remained around 600 nm at daytime (**Figure S5c**), suggesting that BCc could undergo continual aging reactions under relatively lower RH, but produce relatively thinly coated BCc with smaller sizes than those at nighttime (average size of 650 nm). The mean nocturnal WS decreased to 2 m s^{-1}, indicating that the regional atmosphere becomes stagnant (**Figures S5a, b)**. As mentioned earlier and underscored here again, this stagnant and humid atmospheric condition may promote aqueous or heterogeneous reactions, likely further leading to the production of more thickly coated BCc than daytime ones.

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4. Conclusions and implications

During the summer of 2021, the COVID-19 lockdown imposed in Yangzhou resulted in a significant decrease in anthropogenic emissions from traffic and manufacturing sectors. To examine the effects of this lockdown, we utilized spaceborne observations,

ground-based measurements, and particularly SPA-MS analysis to explore the variations, aging characteristics, and sources of BCc in the YRD. We showed that the strict emission controls effectively reduced local gaseous pollutants. However, the decline in NO_x (-39%) and TVOC (-14%) levels might on the other hand result in increased O_3 (28%), leading to a rise in BC-aged particles and a slight elevation in $PM_{2.5}$ levels during the lockdown. Our results revealed a strong non-linear response of $PM_{2.5}$ and O_3 to the gaseous precursors.

The SPA-MS analysis results further demonstrate significant enhancement of OM and SNA coating species on BC-fresh particles, owing to gas-to-particle partitioning and nocturnal multiphase chemistry. Consequently, we observed a higher fraction of BC-aged particles (73%) during the lockdown due to enhanced oxidizing capacity and high relative humidity (RH > 85%). The BC-fresh particles tended to mix with SNA under high RH conditions, while high TVOC levels were accompanied by BCOC formation. However, BCOC particles generally exhibited smaller sizes compared to BC-SNA particles. Moreover, we propose that aqueous or heterogeneous reactions might be important to generate BCOC and BC-SNA particles, especially ones with 400 nm D_{va}, while coagulation might play a more prominent role in larger BC-aged particles. The aging process during LD promoted the conversion of partly coated particles to totally coated ones, with larger diameters (600 nm) and thicker coatings.

It should be noted that the observed average $PM_{2.5}$ concentration during the lockdown in Yangzhou was 20 μg m^{-3}, which still significantly exceeds the WHO's air quality guideline of 5 μg m^{-3}. Our research underscores the crucial role of BCc, which constitutes a significant portion of $PM_{2.5}$, in particulate matter pollution. These particles originate from diverse combustion sources and their behavior is intricately influenced by complex chemistry, regional transport, and meteorological factors. Mere reductions in local primary emissions from traffic and manufacturing sectors exhibit limited efficacy in air quality improvement. Therefore, effective air quality remediation strategies necessitate nuanced control of BCc alongside broader emission reduction efforts. We suggest a more comprehensive regulation of precursor gases from multiple sectors, a wide-ranging joint regulation approach as well as proper consideration of the chemistry, to develop an effective strategy for air quality improvement.

Data availability. The data in this study are available from the corresponding author upon request (caxinra@163.com).

Author contributions. XG, JW, and YD designed the research. YD, HW, and SC conducted the field measurements. YD, HW, JW, and SC analyzed the data. XG, JW, HL, YW, YZ, and EA reviewed the paper and provided useful suggestions. YD, JW, and XG wrote the first draft of the paper. All people were involved in the discussion of the results.

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Competing interests. The contact author has declared that neither they nor their co-authors have any competing interests.

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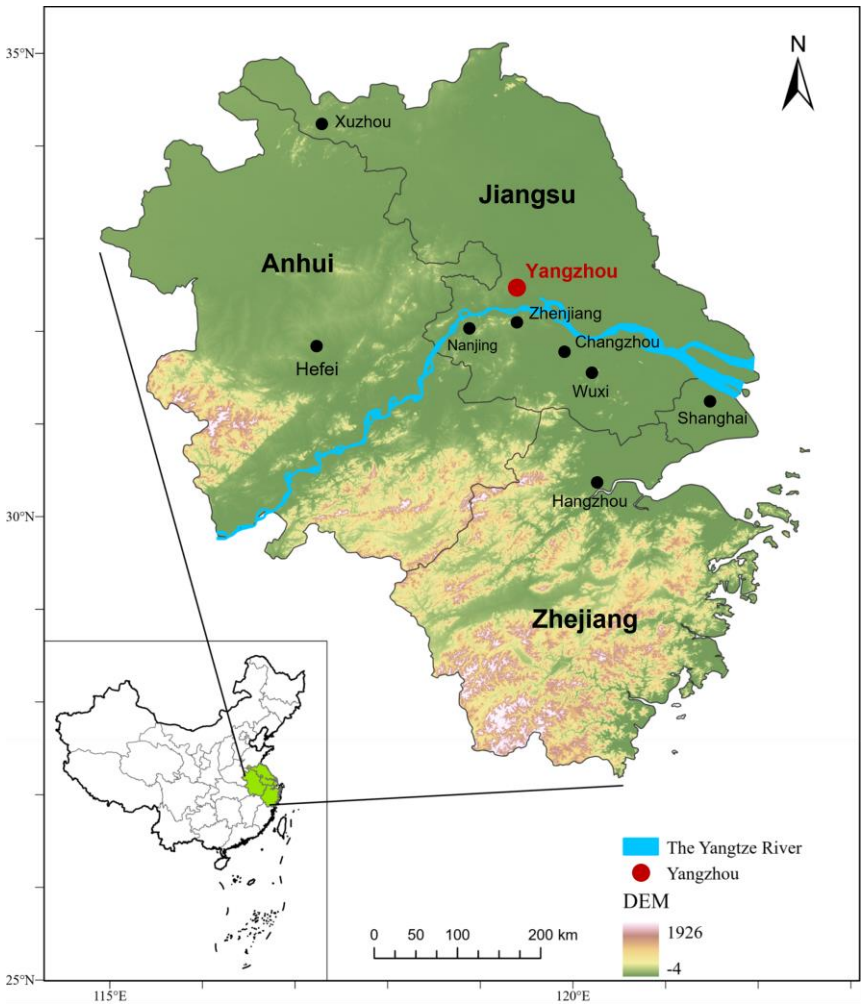


Figure 1. Geographical overview of the Yangtze River Delta (YRD) Region in China,

depicting the major cities within the YRD and the sampling site located in Yangzhou. The color gradient from green to white indicates varying altitudes across the region (Maps were generated by using ArcGIS Pro).

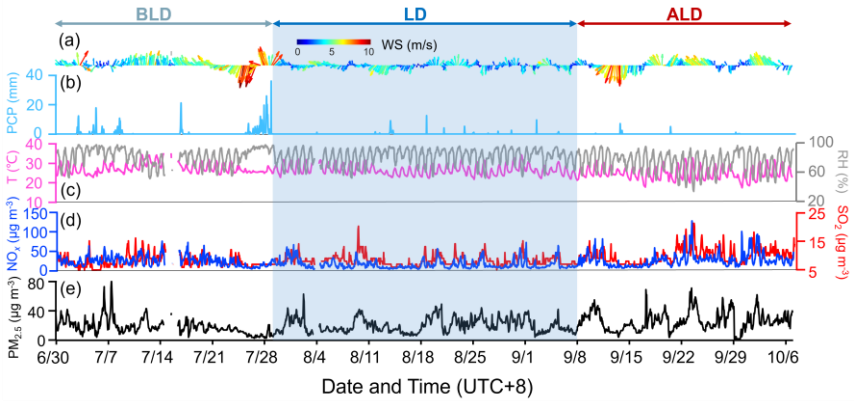


Figure 2. Temporal variations of **(a)** wind direction (WD) and wind speed (WS), **(b)** precipitation (PCP), **(c)** temperature (T) and relative humidity (RH), **(d)** concentrations of NO_x and SO_2, and **(e)** mass loading of $PM_{2.5}$. The grey, blue, and red arrow ranges denote the periods before lockdown (BLD), during lockdown (LD), and after lockdown (ALD).

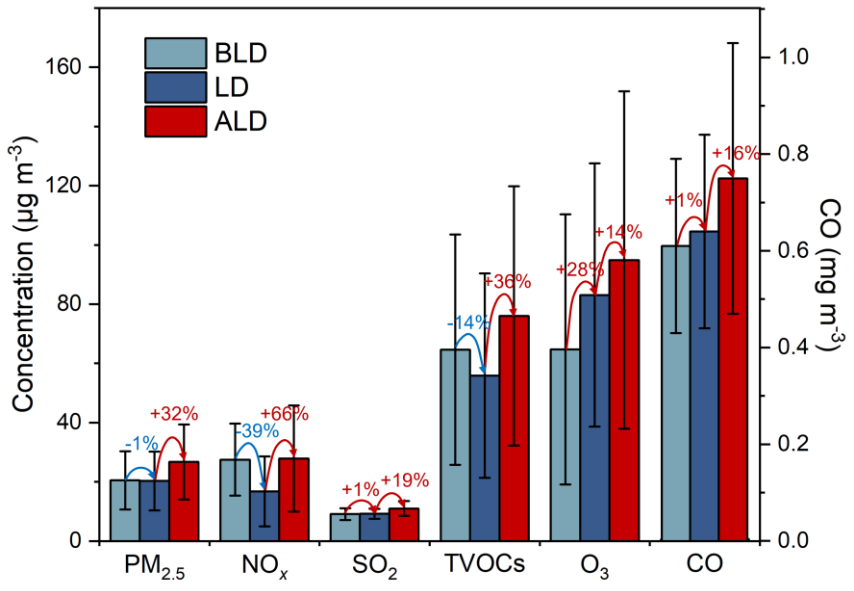


Figure 3. Ground-based observations of $PM_{2.5}$, NO_x, SO_2, O_3, CO, and TVOC

concentrations in Yangzhou. The figure compares the averages during the BLD (grey), LD (blue), and ALD (red) periods. Error bars indicate SDs over different lockdown periods.

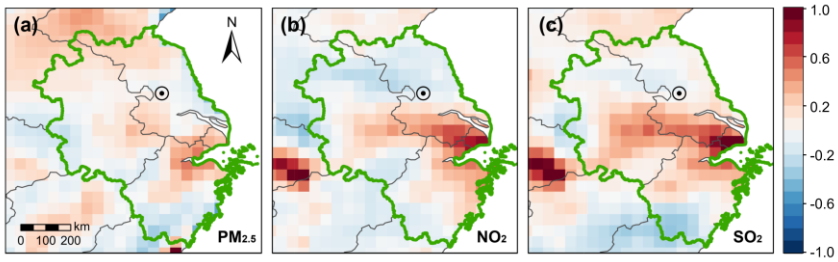


Figure 4. The fractional changes (i.e., (LD – BLD)/BLD) of **(a)** $PM_{2.5}$, **(b)** NO_2, and **(c)** SO_2 between BLD and LD periods based on spaceborne measurement. The circle symbols in the maps indicate the location of Yangzhou, and the green region represents the YRD.

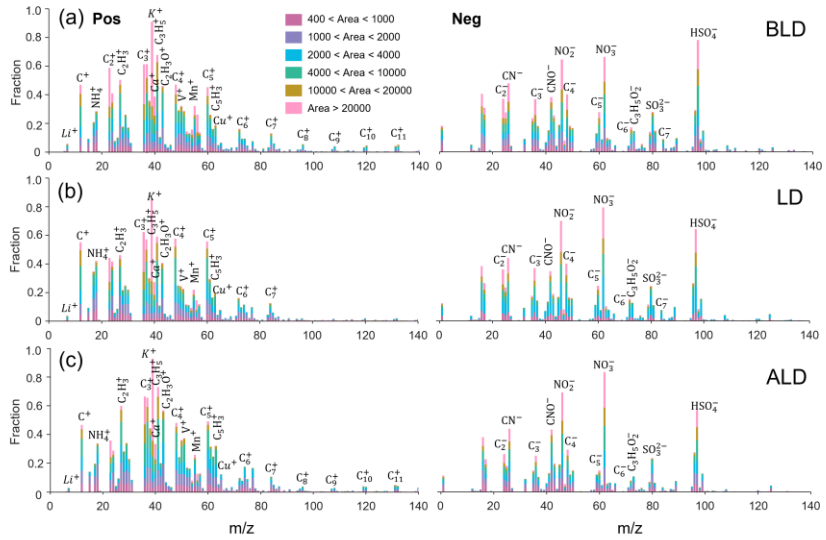


Figure 5. The average positive and negative mass spectra of BCc **(a)** before the lockdown period (BLD), **(b)** during the lockdown period (LD), and **(c)** after the lockdown period (ALD).

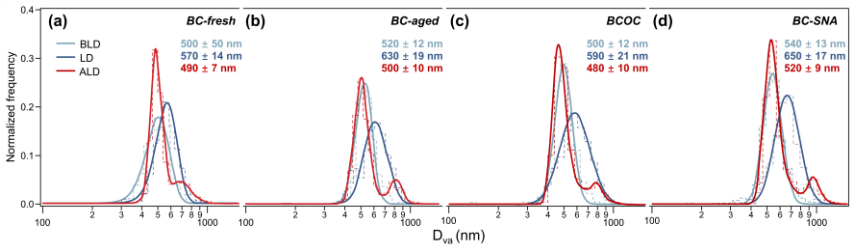


Figure 6. Size distribution of different types of BCc during different periods in Yangzhou. **(a)** BC-fresh particles, **(b)** BC-aged particles, **(c)** BCOC particles, and **(d)** BC-SNA particles. The Log-normal distribution was used to fit the unimodal size distribution, and the Lorentz distribution was used to fit the bimodal size distribution. The corresponding mode sizes (with the standard deviations) are also shown.

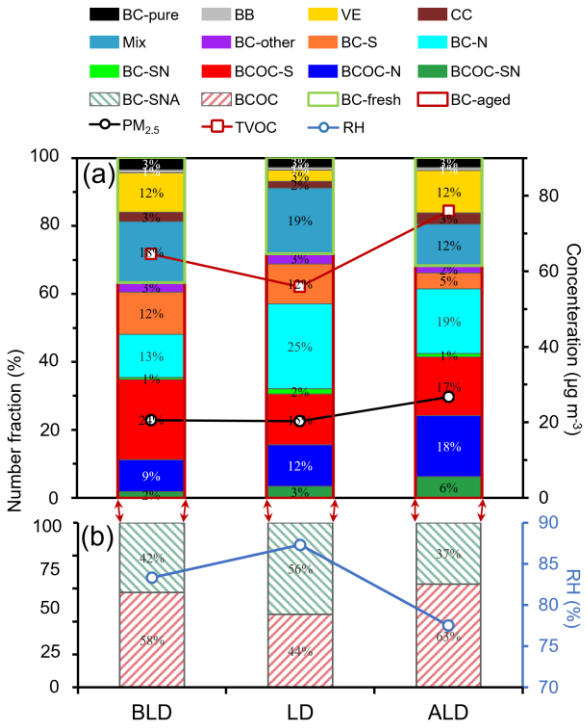


Figure 7. Number fractions of BCc. **(a)** The number fractions of different BCc along with the concentrations of $PM_{2.5}$ and total volatile organic compounds (TVOC). **(b)** The

number fractions of different types of BC-aged particles along with relative humidity (RH).

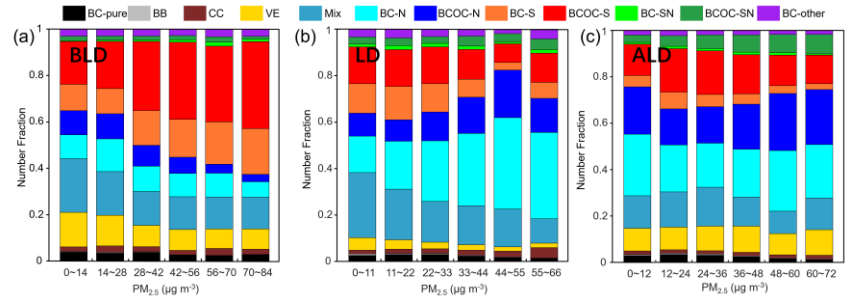


Figure 8. Variations of number fractions of BCc particle types with $PM_{2.5}$ mass concentrations during **(a)** the BLD period, **(b)** LD, and **(c)** the ALD period.

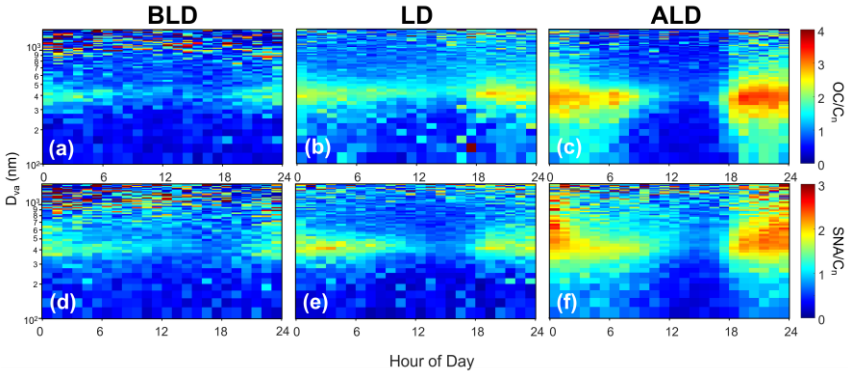
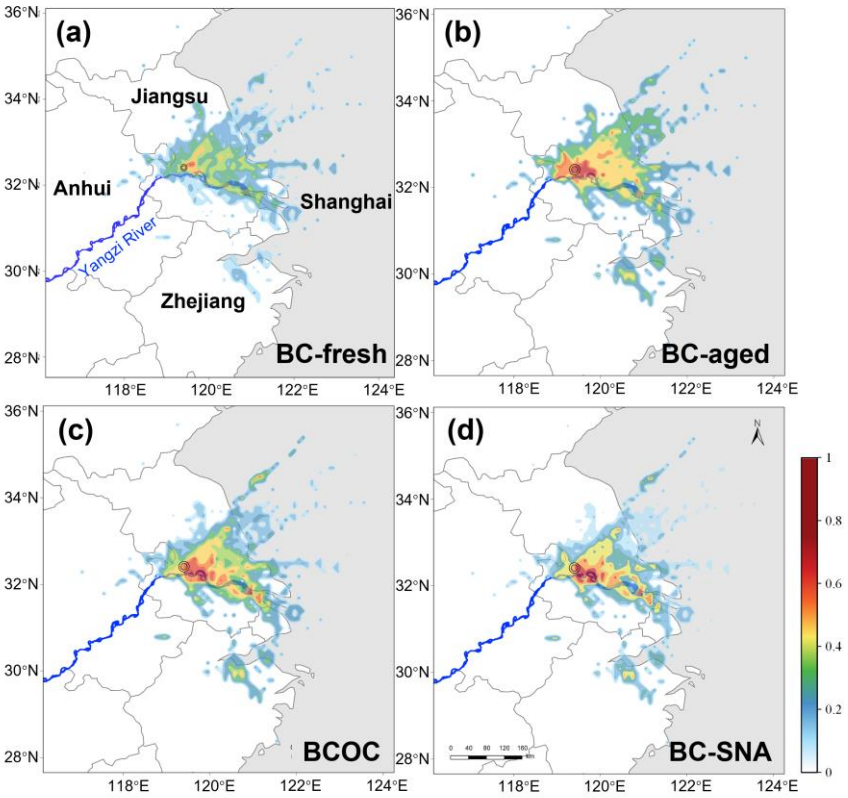


Figure 9. Diurnal variations of the ratios of OC/Cn and SNA/Cn with a size distribution of BCc during **(a, d)** BLD, **(b, e)** LD, and **(c, f)** ALD.



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1026 **Figure 10.** The PSCF maps for different BCc during LD. **(a)** BC-fresh. **(b)** BC-aged.
1027 **(c)** BCOC. **(d)** BC-SNA.
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