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 $\leq$ 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

86 (Wang et al., 2010; Zhou et al., 2010), the 2015 Asia-Pacific Economic Cooperation
87 (APEC) (Zhu et al., 2015), the 2014 Nanjing Youth Olympic Games (Wang et al., 2022)
88 and the national COVID-19 lockdown in 2020 winter (Huang et al., 2021; Le et al.,
89 2020; L. Li et al., 2020; Wang et al., 2020). Previous studies extensively investigated
90 air pollutant variations during the COVID-19 lockdown in the winter of 2020 across
91 different regions of the world. Stringent restrictions on industrial and vehicular
92 activities have resulted in significant reductions in gaseous pollutants and particulate
93 matter, not only in megacities (Chen et al., 2020; Jeong et al., 2022; Sun et al., 2020)
94 but also in middle-sized cities (Clemente et al., 2022; Wang et al., 2021; Xu et al., 2020)
95 and rural areas (Cui et al., 2021, 2020; Jain et al., 2021). Compared to the decreasing
96 trends observed in most cities worldwide, the level of $PM_{2.5}$ in Shanghai (Chang et al.,
97 2020), Hohhot (Zhou et al., 2022), and the Northeast of China Plain (Nie et al., 2021)
98 increased unexpectedly. These observations reveal the complex aerosol chemistry of
99 $PM_{2.5}$ comprising primary and secondary components. The reduction of primary
100 pollutants during lockdown resulted in a shift towards a higher proportion of secondary
101 aerosols, including inorganic and organic species, exhibiting a non-linear response to
102 emission changes (Zhang et al., 2021). Furthermore, some studies suggested that the
103 increase in secondary aerosols during lockdown is due to the enhanced atmospheric
104 oxidative capacity resulting from the rise in ozone levels (Wang et al., 2021),
105 unfavorable meteorological conditions (Chien et al., 2022; Sulaymon et al., 2021a),
106 changes of local and regional emission sources (Feng et al., 2022). However, most
107 previous studies focused on lockdown events during the cold seasons, and studies on
108 summer lockdown events in China were very limited.
109
110 Yangzhou is located in the central region of the Yangtze River Delta (YRD), at the
111 junction of the Yangtze River and, the Beijing-Hangzhou Grand Canal, which serves as
112 a prominent economic city, industrial-intensive area, and highly active inland shipping
113 node in East China. Due to the complex emissions and feedback with the East Asian
114 monsoons (Ding et al., 2019), this region is susceptible to anthropogenic aerosols,
115 especially BCc originating from chemical, steelmaking, coal-fired, petrochemical
116 enterprises, and transportation, etc. Extensive studies have investigated the responses
117 of atmospheric pollutants to emission changes during the COVID-19 lockdown
118 measures in the YRD (Chen et al., 2021; Li et al., 2020; Qin et al., 2021; Zhang et al.,
119 2022). However, the key chemical and physical processes specifically responsible for
120 the BCc in this region are still unclear. During the summer of 2021, Yangzhou
121 experienced a resurgence of COVID-19 with over 500 confirmed cases. In response,
122 stringent public health measures were imposed from July 29th to September 10th,
123 including the closure of public transport, and suspension of non-essential industrial
124 plants, restaurants, shopping malls, and entertainment clubs. People were also
125 mandated to quarantine at home. Consequently, Yangzhou experienced a significant
126 decline in transportation and industrial energy consumption, dropping by nearly 46%
127 and 25%, respectively, compared to the same period in 2020 (www.yangzhou.gov.cn),

128 implying a substantial reduction in human activity and primary emissions. Unlike the
129 nationwide COVID-19 lockdown in China during the cold season of 2020 (Le et al.,
130 2020; Sulaymon et al., 2021b), the summer lockdown in Yangzhou was more localized
131 but protracted, significantly altering local anthropogenic emissions while neighboring
132 cities maintained regular operations, which provides a unique opportunity to explore
133 and compare the diverse mixing states and, the aging process of BCc in different
134 anthropogenic emission conditions in summer. Here we report the chemical
135 compositions and aging characteristics of airborne BCc in YRD. Our investigation
136 involved a combination of ground measurements, spaceborne observations, and mass
137 spectrometric analysis conducted during the COVID-19 lockdown in the summer of
138 2021 in Yangzhou. Additionally, we employed potential source contribution function
139 (PSCF) analysis to investigate the air pollution patterns in the YRD.
140

2. Methods

2.1 Sampling site and instruments

143 The in-situ online measurements were conducted at a rooftop laboratory 20 m above
144 ground located in a national air quality monitoring station, Yangzhou Environmental
145 Monitoring Center (32.41ºN, 119.40ºE), Yangzhou, China (**Fig. 1**). This sampling site
146 is a typical urban site surrounded by residential areas, arterial roads, parks, restaurants,
147 and shopping centers. In this study, the measurement period was divided into three
148 phases: the before-lockdown period (BLD: 30 June to 28 July 2021), the lockdown
149 period (LD: 29 July to 9 September 2021), and the after-lockdown period (ALD: 10
150 September to 7 October 2021) (**Fig. 2**).
151

152 A single-particle aerosol mass spectrometer (SPA-MS, Hexin Analytical Instrument Co.,
153 Ltd., China) was deployed during the field campaign to obtain the chemical
154 composition, size distribution, and mixing state of individual $PM_{2.5}$ particles. A cyclone
155 with a 2.5 μm cutpoint (Model URG-2000-30ED) and a Nafion dryer is equipped in
156 front of the sampling inlet. Individual particles are introduced into the SPA-MS through
157 a critical orifice at a flow rate of 3 L min^{-1}. The vacuum aerodynamic diameters (D_{va})
158 are determined using the velocities derived from two continuous laser beams (diode Nd:
159 YAG, 532 nm) spaced 6 cm apart. Subsequently, these particles are desorbed and
160 ionized by a downstream pulsed laser (266 nm), and ion fragments are generated and
161 measured by a Z-shaped bipolar time-of-flight mass spectrometer. A more detailed
162 description of SPA-MS can be found in previous studies (Li et al., 2011).
163

164 $PM_{2.5}$ mass concentration was measured by a particulate matter monitor (XHPM2000E,
165 Xianhe, China). Nitrogen oxides ($NO_x = NO + NO_2$), SO_2, and ozone (O_3)
166 concentrations were detected with a set of Thermo Fisher Scientific instruments
167 (Models 42i, 43i, and 49i). The concentrations of 103 volatile organic compounds
168 (VOCs) in ambient air, comprising 57 ozone precursors (PAMS), 12 aldehydes and
169 ketones, and 34 toxic organics (TO15), were continuously monitored at hourly intervals

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

177 2.2 Data analysis
178 **2.2.1 Satellite Product**
179 In this study, we utilized the Copernicus Atmosphere Monitoring Service (CAMS)
180 Global Near-Real-Time dataset (available at https://developers.google.com/earth-
181 engine/datasets/catalog/ECMWF_CAMS_NRT), acquired from the European Centre
182 for Medium-Range Weather Forecasts (ECMWF), to analyze the distribution of total
183 surface column concentrations of NO_2, SO_2 and surface $PM_{2.5}$ mass concentration.
184 CAMS offers the capacity to continuously monitor the composition of the Earth's
185 atmosphere at global and regional scales since 2016, with a spatial resolution of 44528
186 meters (Benedetti et al., 2009; Morcrette et al., 2009). The details of the bands of the
187 dataset used in this study are shown in Table S2. We calculated and plotted the averaged
188 2-dimensional data of ECMWF/CAMS/NRT NO_2, SO_2, and $PM_{2.5}$ during BLD and LD
189 over the region of interest (17.93~54.74 ºN, 71.21~142.23 ºE) using Google Earth
190 Engine (Gorelick et al., 2017). The integration of remote sensing measurements has
191 provided a more comprehensive understanding of the sources and distributions of
192 particle matter and gaseous pollutants facilitating the evaluation of the impact of human
193 activities on air quality.

194 **2.2.2 Geographic Source Analysis**

195 The potential source contribution function (PSCF) analysis, based on the Hybrid
196 Single-Particle Lagrangian Integrated Trajectory (HYSPLIT) model, can be employed
197 to identify regional sources of air pollutants. Before conducting the PSCF analysis, 36
198 hours of air mass backward trajectories with one-hour resolution at 500 m above ground
199 level were calculated using the wind data from the Global Data Assimilation System
200 (GDAS) provided by the National Oceanic and Atmospheric Administration (NOAA)
201 (Wang et al., 2009). An open-source software MeteoInfo (Wang, 2014) was utilized for
202 the PSCF analysis. The whole study area (110.1~133.4 ºE and 21.3~39.9 ºN) covered
203 by the trajectories was divided into thousands of cells with a spatial resolution of 0.1°
204 × 0.1°. The PSCF was simulated according to the following equation:

205
$$PSCF_{ij} = \frac{m_{ij}}{n_{ij}} \qquad (1)$$

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

210 percentile of hourly average number fractions, respectively. To further improve the
211 accuracy of the PSCF analysis and minimize analytical uncertainties, the Weighted
212 PSCF (WPSCF) functions as shown in Equation (2~3) were applied (Polissar et al.,
213 1999). The weight (W_{ij}) for each grid cell was determined based on the number of
214 trajectory endpoints (n_{ij}) as follows:

$$WPSCF_{ij} = W_{ij} \times PSCF_{ij} \tag{2}$$

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

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

2.2.3 SPA-MS Data Analysis

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

3. Results and discussion

3.1 Field observations

231 **Figure 2** presents the temporal variations of meteorological parameters, PM$_{2.5}$, NO$_x$,
232 and SO$_2$ concentrations. Notably, significantly reductions in PM$_{2.5}$, NO$_x$, and SO$_2$ were
233 observed at the end of BLD due to a high precipitation event, with a peak hourly
234 precipitation reaching 37 mm, and the data collected during this event were excluded
235 from the analysis. During BLD, the mean temperature (T) was 28±3 °C, the total
236 precipitation was 221 mm, with an average relative humidity (RH) of 83±12%. The
237 prevailing winds originated from the south and southeast, with a mean wind speed (WS)
238 of 3.3±1.2 m s^{-1}. In comparison, LD shows a decline in temperature to 27±2 °C and WS
239 to 2.3±1.0 m s^{-1}, but an increase in RH to 87±11% and a reduction in total precipitation
240 to 86 mm. **Figure S2b and c** present uniform distributions of RH and boundary-layer
241 height (BLH) across the YRD during LD. These regional meteorological conditions and
242 the effective removal of the pollutants accumulated at the end of BLD facilitated the
243 investigation of BCc regional transport in YRD. During ALD, the temperature declined
244 further to 25±3 °C, WS increased to 3.3±1.5 m s^{-1}, and total precipitation dropped to 27
245 mm with a lower RH of 77±14%.

247   During LD, strict measures resulted in notably lower surface concentrations of $PM_{2.5}$
248   (20.3 μg m^{-3}), NO_x (16.8 μg m^{-3}) and TVOC (55.9 μg m^{-3}) compared to BLD and ALD.
249   Conversely, the surface O_3 concentration showed an increase of 18.4 μg m^{-3} (28%)
250   during LD relative to BLD. The reduction of fresh NO emission alleviates O_3 titration
251   (Steinfeld, 1998) could be an explanation. Analysis from **Fig. S3** indicates that the O_3
252   level is higher than those of neighboring cities in the YRD, suggesting higher local
253   atmospheric oxidation capacity during LD. However, the average concentrations of
254    $PM_{2.5}$ (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})
255   were comparable during both BLD and LD (**Fig. 3**). After LD, social activities
256   gradually resumed in Yangzhou City, leading to an apparent increase in all observed
257   pollutants during the ALD period. For instance, there were relative increases of 66%
258   for NO_x, 19% for SO_2, 36% for TVOC, 14% for O_3, 32% for $PM_{2.5}$, and 16% for CO
259   from LD to ALD, respectively (**Fig. 3**).
260
261   Satellite-retrieved $PM_{2.5}$, NO_2, and SO_2 data over the entire region of eastern China
262   were also investigated, and results show that these pollutants were predominantly
263   concentrated in Shanghai and its neighboring cities, including Yangzhou, during both
264   BLD and LD (**Fig. S4**). **Figure 4** presents regional fractional changes of mean $PM_{2.5}$,
265   NO_2, and SO_2 concentrations from the BLD to LD periods in YRD, all showing an
266   increase of 29%, 6%, and 14%, respectively. In comparison, Yangzhou city experienced
267   lower increases in these air pollutants, with slight changes of 6%, -18%, and -4% for
268    $PM_{2.5}$, NO_2, and SO_2, respectively. The implication is that, even though local primary
269   emissions, such as NO_2, and SO_2, were reduced substantially during LD, they still could
270   be affected by regional transport. Furthermore, as depicted in **Fig. S3**, the
271   concentrations of NO_2 in major cities of the YRD were more than twice higher than in
272   Yangzhou during LD, confirming a relatively lower local primary emissions due to the
273   stringent lockdown. However, the higher level of SO_2 in Yangzhou during LD may be
274   attributed to the nearby power stations along the Yangtze River, which were not
275   impacted by the lockdown measures.
276
277   ## 3.2 Chemical composition and size distribution of individual BCc
278   Based on the SPA-MS analysis, a total of 1068362 BCc was collected during the whole
279   study period. The BCc accounted for 59%, 69%, and 57% of the total number of
280   measured particles in the BLD, LD, and ALD periods, respectively. **Figure 5** shows the
281   normalized average mass spectra of BCc during three periods. Ion height in each
282   spectrum reflects the number fraction of the detected BCc with the corresponding ion
283   to the total BCc, while colors represent peak area ranges of detected ions. BCc in BLD,
284   LD, and ALD shown similar mass spectra at m/z < 100, with common peaks including
285   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]$^+$,
286   63[C_5H_3]$^+$, 46[NO_2]$^-$, 62[NO_3]$^-$, and 97[HSO_4]$^-$. However, the abundance of large m/z
287   carbon ions ($C_n^{\pm}$, n > 7) in both BLD and ALD periods was 1.5 times higher than that in
288   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]$^+$, 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

331   types are shown in **Table S1** and **Fig. S1** in the Supplement.

332

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

342

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

362

363   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
364   fraction of BC-fresh particles rose from 28% (LD) to 31% (ALD), while the fraction of
365   VE particles also increased from 3% (LD) to 12% (ALD) (**Fig. 7a**), coinciding with a
366   16% rise in CO concentration. This suggests a shift in combustion practices and fuel
367   usage patterns as economic activities resumed. The increased CO levels, a known
368   marker for combustion-related emissions, align with the resurgence of vehicle
369   emissions and other activities that emit BC and CO concurrently (Wang et al., 2015,
370   Zhou et al., 2009). Notably, the size distributions of BC-fresh and BC-aged particles
371   presented relatively small peaks at 690 nm and 820 nm during ALD, in addition to the
372   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 (**Fig. 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 (**Fig. 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 (**Fig. 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 (**Fig. 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 (**Fig. 8a**), whereas BC-N played a vital role in the $PM_{2.5}$ increase during LD (**Fig. 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 (**Fig. 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-

415   N during LD might be attributed to regional transport, similar to that in Shanghai during
416   2020 winter lockdown (Chang et al., 2020).

417

418   ## 3.3   Chemical aging of BCc

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

434

435   **Figure 9** illustrates the diurnal variations of the OC/C_n and SNA/C_n ratios along with
436   the size distribution of BCc during different periods. We observed that both OC/C_n and
437   SNA/C_n increased during nighttime and decreased during daytime. These variations
438   showed the prominent enhancements of nocturnal OM and SNA, which could be
439   attributed to the accelerated gas-to-particle partitioning and nocturnal secondary
440   formation of organic/inorganic components under high relative humidity (RH > 85%)
441   and relatively stagnant air mass (WS < 3 m s^{-1}) (**Fig. S5**). It is worth noting that from
442   BLD to LD and ALD, the intensity of diurnal variations of OC/C_n and SNA/C_n
443   increased obviously. This discrepancy can be attributed to several reasons. (i) During
444   BLD, the frequent precipitations effectively scavenged the particles (Isokääntä et al.,
445   2022); (ii) In contrast, stronger solar radiation and higher O_3 concentration during LD
446   promoted photochemical formations of OC and SNA; (iii) After lockdown, more
447   precursors due to increased local emissions may lead to more production of secondary
448   components than that during BLD as explained earlier. These results indicate that the
449   aging process and mixing state of BCc depend strongly on meteorological conditions
450   as well as emission sources in urban cities.

451

452   As shown in **Fig. 9**, BCc with ~400 nm D_{va} exhibited significant diurnal fluctuations in
453   the OC/C_n and SNA/C_n ratios, during LD. There is a noticeable increase in the
454   proportion of BC-SNA particles during nighttime when RH is relatively high. These
455   observations suggest that nighttime heterogeneous hydrolysis may be considered a key
456   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 (**Fig. 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) (**Fig. 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.

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 **Fig. 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 (**Fig. 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 **Fig. 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 (**Fig. 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 (**Fig. 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.

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},

541 while coagulation might play a more prominent role in larger BC-aged particles. The
542 aging process during LD promoted the conversion of partly coated particles to totally
543 coated ones, with larger diameters (600 nm) and thicker coatings.
544
545 It should be noted that the observed average $PM_{2.5}$ concentration during the lockdown
546 in Yangzhou was 20 μg m^{-3}, which still significantly exceeds the WHO's air quality
547 guideline of 5 μg m^{-3}. Our research underscores the crucial role of BCc, which
548 constitutes a significant portion of $PM_{2.5}$, in particulate matter pollution. These particles
549 originate from diverse combustion sources and their behavior is intricately influenced
550 by complex chemistry, regional transport, and meteorological factors. Mere reductions
551 in local primary emissions from traffic and manufacturing sectors exhibit limited
552 efficacy in air quality improvement. Therefore, effective air quality remediation
553 strategies necessitate nuanced control of BCc alongside broader emission reduction
554 efforts. We suggest a more comprehensive regulation of precursor gases from multiple
555 sectors, a wide-ranging joint regulation approach as well as proper consideration of the
556 chemistry, to develop an effective strategy for air quality improvement.

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

559

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

565

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571

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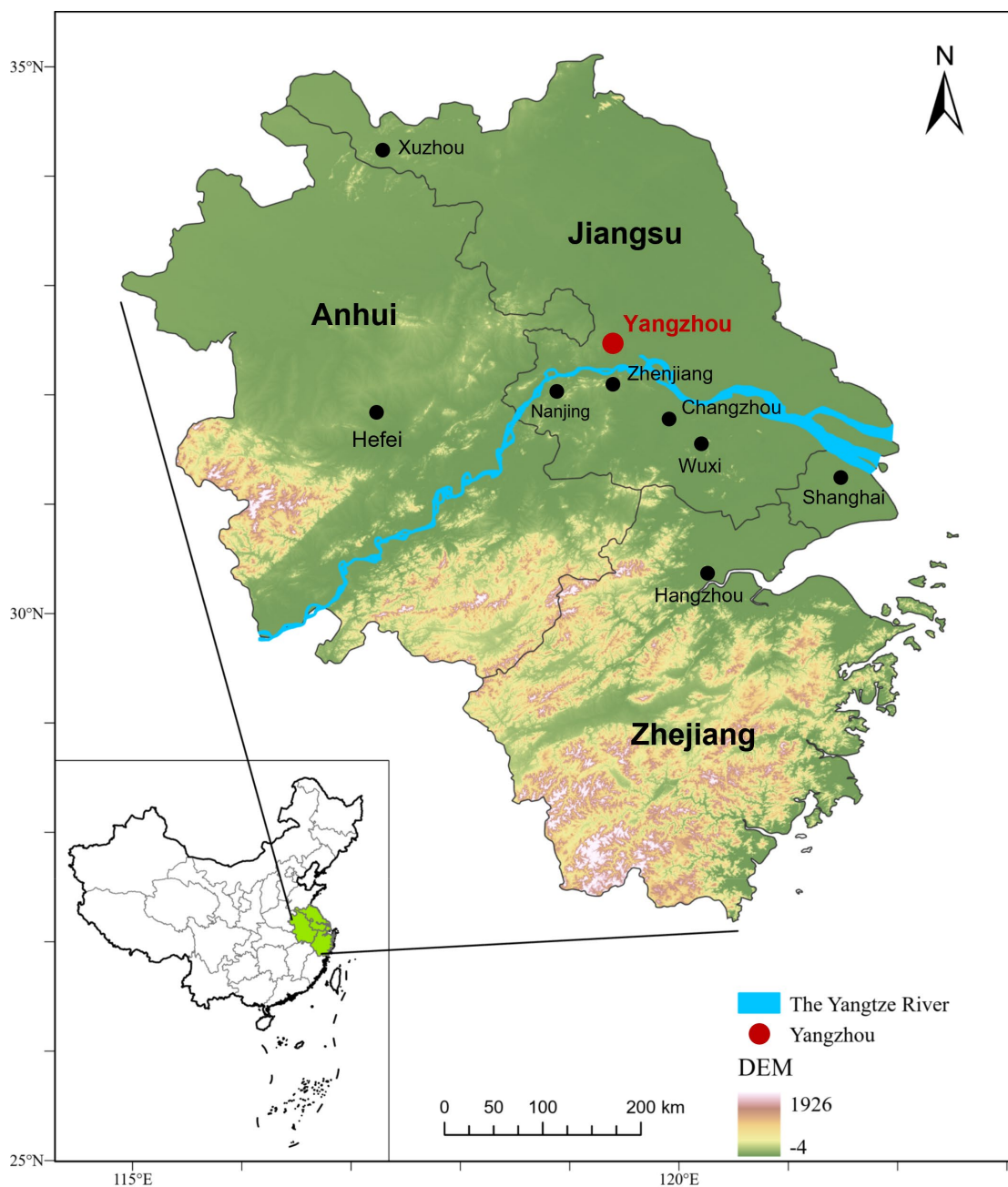
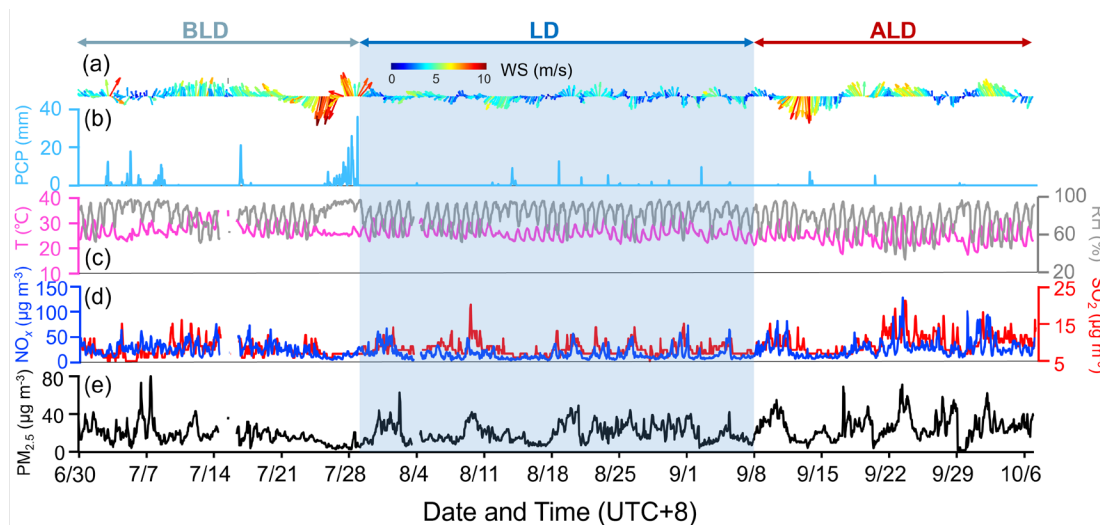
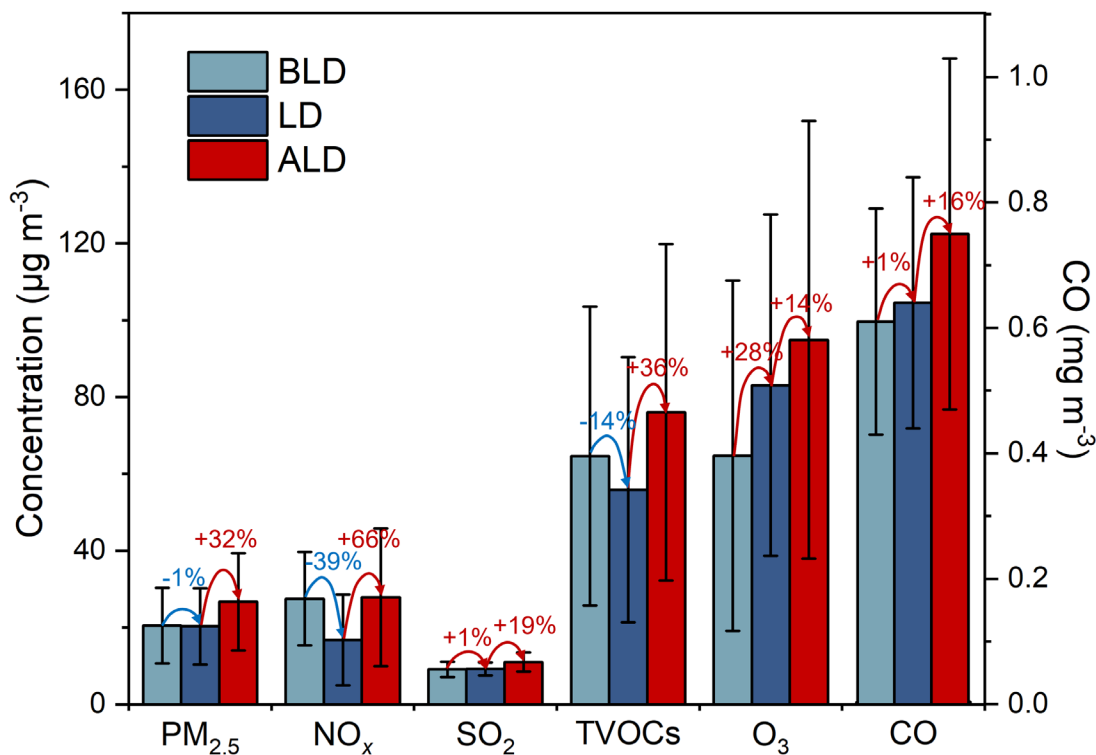


Figure 1. Geographical overview of the Yangtze River Delta (YRD) Region in China,

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

969

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

975

976 **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.

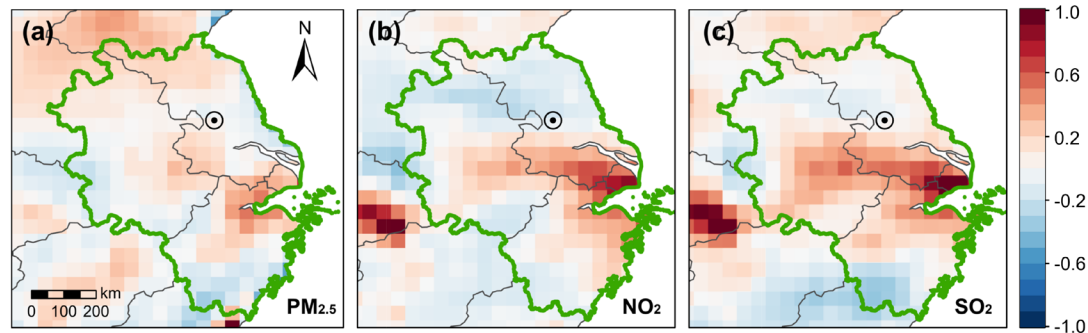



Figure 4. The fractional changes (i.e., (LD – BLD)/BLD) of **(a)** $PM_{2.5}$, **(b)** NO_2, and **(c)** SO_2 between BLD and LD periods based on spaceborne measurement. The circle symbols in the maps indicate the location of Yangzhou, and the green region represents the YRD.

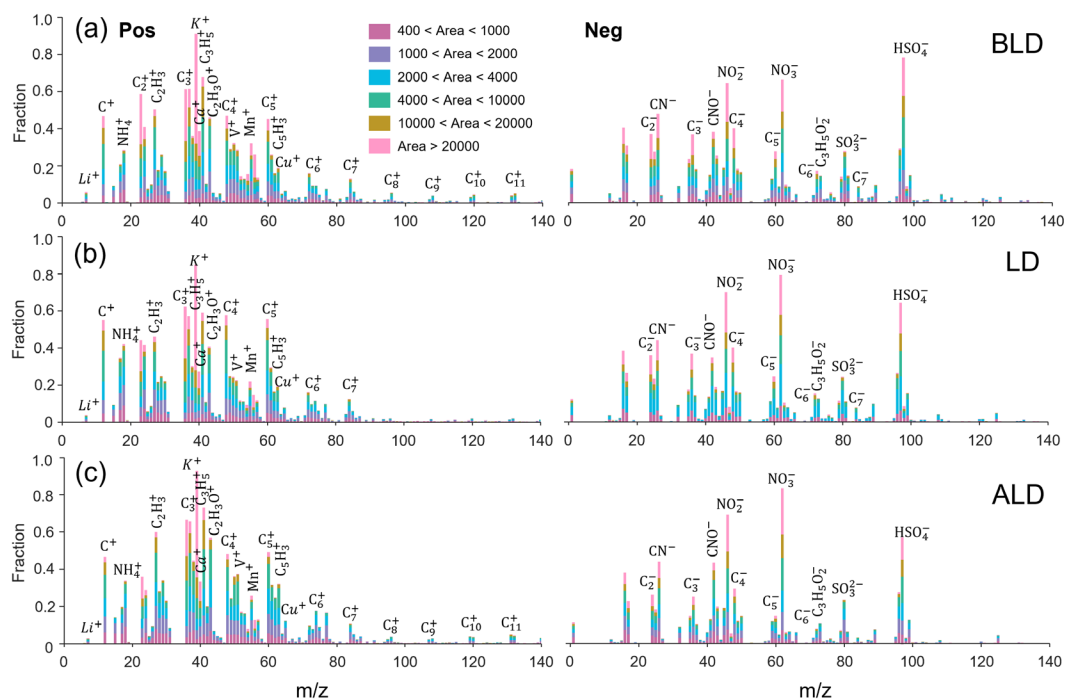



Figure 5. The average positive and negative mass spectra of BCc **(a)** before the lockdown period (BLD), **(b)** during the lockdown period (LD), and **(c)** after the lockdown period (ALD).

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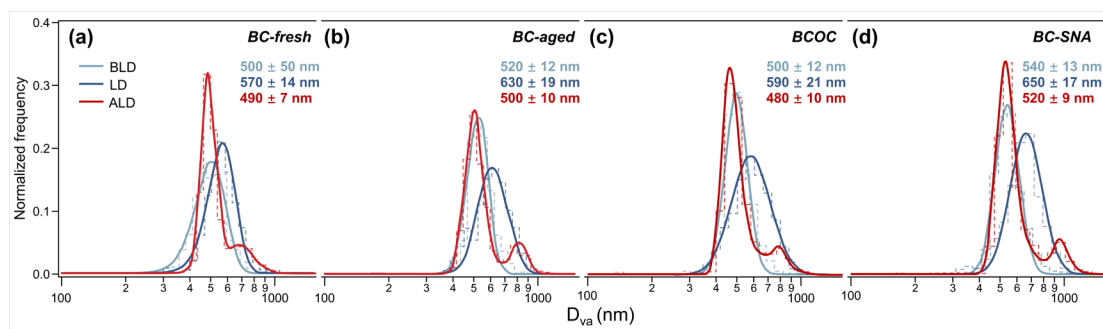
992 **Figure 6.** Size distribution of different types of BCc during different periods in
993 Yangzhou. **(a)** BC-fresh particles, **(b)** BC-aged particles, **(c)** BCOC particles, and **(d)**
994 BC-SNA particles. The Log-normal distribution was used to fit the unimodal size
995 distribution, and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

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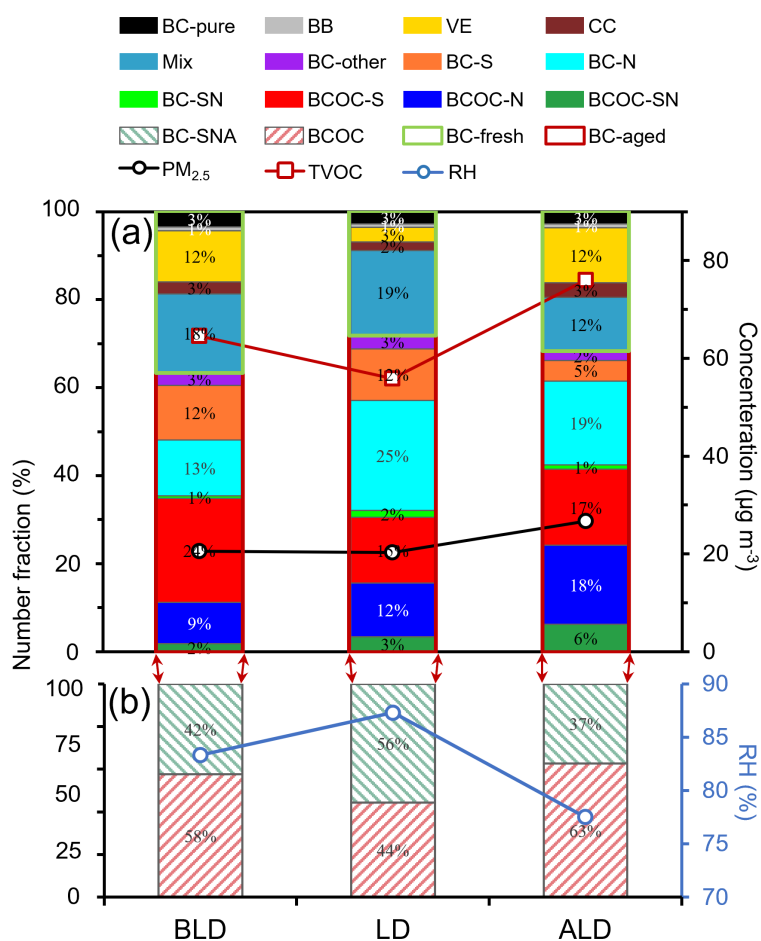
999 **Figure 7.** Number fractions of BCc. **(a)** The number fractions of different BCc along
1000 with the concentrations of $PM_{2.5}$ and total volatile organic compounds (TVOC). **(b)** The

1001 number fractions of different types of BC-aged particles along with relative humidity
1002 (RH).
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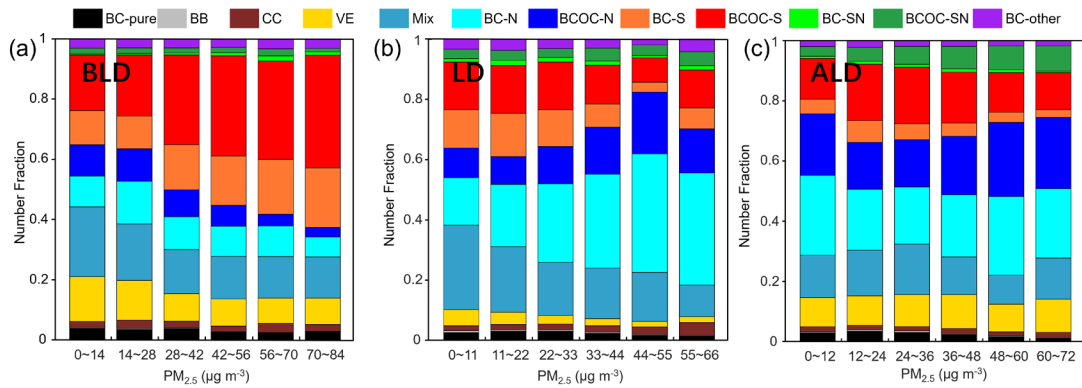
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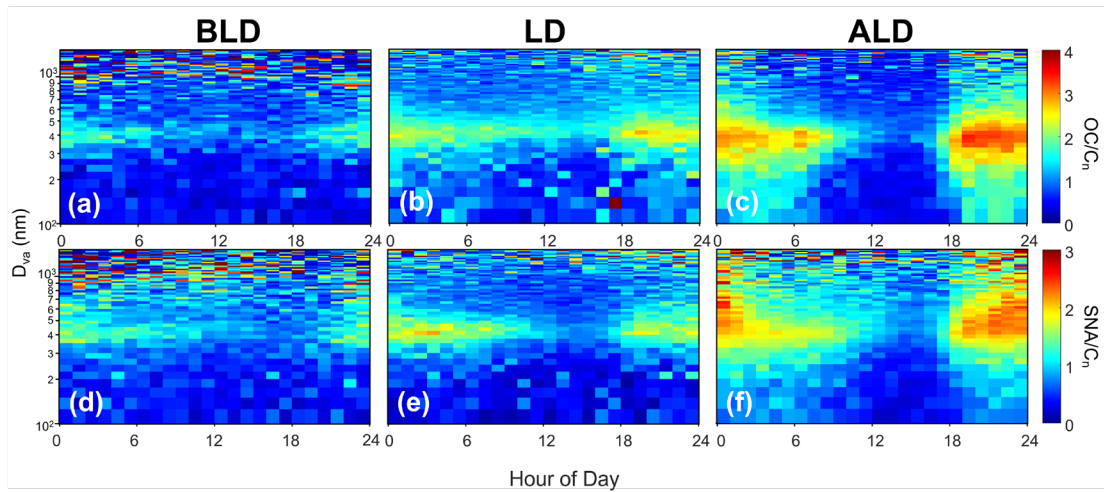
1005 **Figure 8.** Variations of number fractions of BCc particle types with PM$_{2.5}$ mass
1006 concentrations during **(a)** the BLD period, **(b)** LD, and **(c)** the ALD period.

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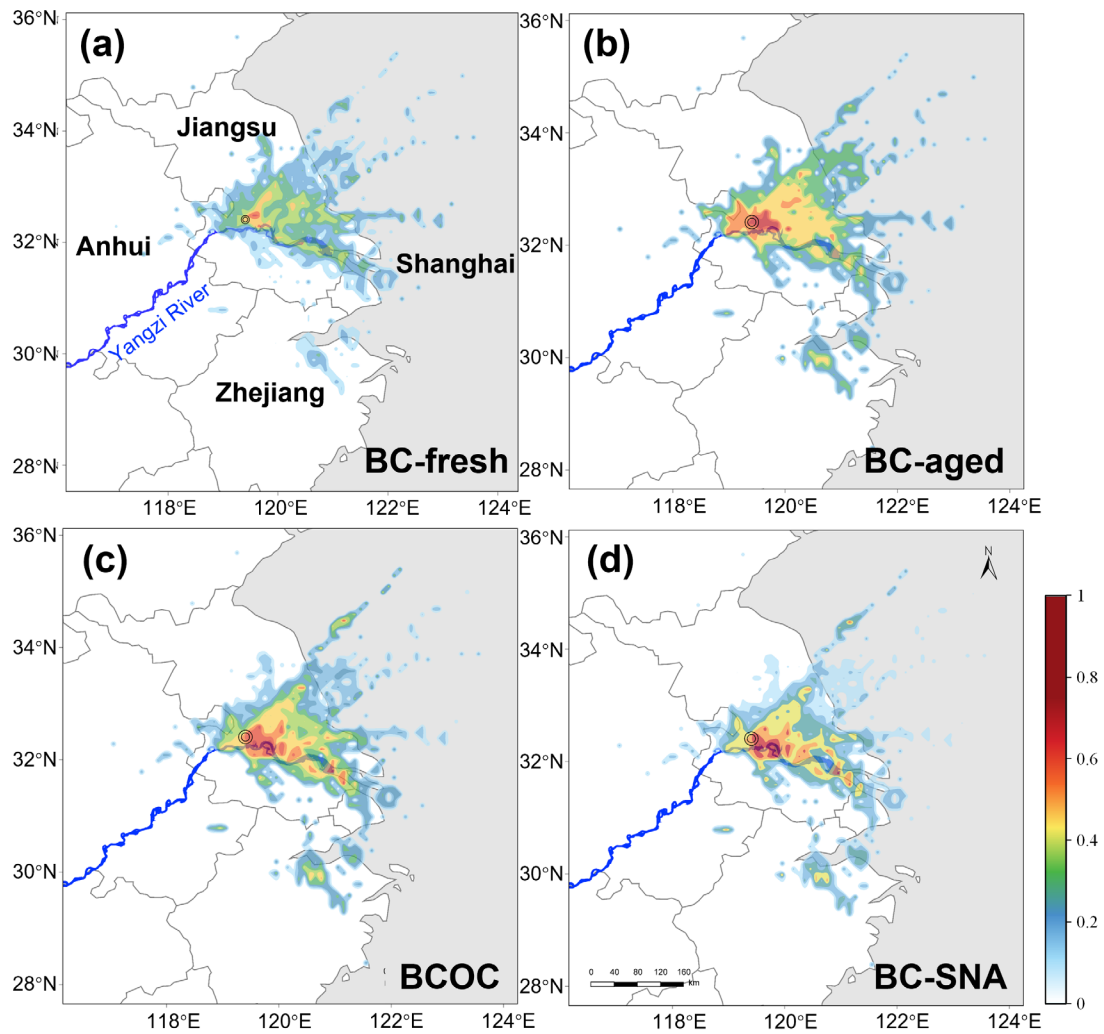
1008 **Figure 9.** Diurnal variations of the ratios of OC/Cn and SNA/Cn with a size distribution
1009 of BCc during **(a, d)** BLD, **(b, e)** LD, and **(c, f)** ALD.

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1011 **Figure 10.** The PSCF maps for different BCc during LD. **(a)** BC-fresh. **(b)** BC-aged.
1012 **(c)** BCOC. **(d)** BC-SNA.

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