



# Measurement report: Characteristics of airborne black carbon-containing particles during the 2021 summer COVID-19 lockdown in Yangzhou, China

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#### 23 Abstract

24 Black carbon-containing (BCc) particles are pervasive in ambient atmosphere. The 25 unexpected outbreak of the COVID-19 pandemic in 2021 summer prompted a localized 26 and prolonged lockdown in Yangzhou City, situated in the Yangtze River Delta (YRD) 27 region, China, which provides a unique opportunity to gain insights into the relationship 28 between emission sources and BCc. Satellite and ground-level measurements both 29 demonstrated that strict emission controls effectively reduced local gaseous pollutants. 30 Meanwhile, single particle aerosol mass spectrometer (SPA-MS) analysis showed that 31 the number fraction of freshly emitted BCc particles decreased to 28% during the 32 lockdown (LD), with that from vehicle emissions experiencing the most substantial 33 reduction. However, the uncontrolled reductions of nitrogen oxides  $(NO_x)$  and volatile 34 organic compounds (VOCs) likely contributed to increased ozone  $(O_3)$  concentrations, 35 increased the oxidizing capacity, which may in turn enhanced secondary PM2.5 formation and compensated the primary PM2.5 reduction. As a result, we did observe a 36 37 slight increase of PM<sub>2.5</sub> concentration (21.2 µg m<sup>-3</sup>) during the LD period compared to 38 the period before the lockdown (BLD), and the increase of more aged BCc particles. 39 Reactive trace gases (e.g., NO<sub>x</sub>, SO<sub>2</sub>, and VOCs) could form thick coatings on pre-40 existing particles likely via enhanced heterogeneous hydrolysis under high RH as well, 41 resulting in significant BCc particle growth (~600 nm) during LD period. Furthermore, 42 BCc source apportionment reveals that BCc particles were primarily of local origin 43 (78%) in Yangzhou during normal summertime. However, coal combustion (23%) and 44 vehicle emissions (21%) were prominent non-local pollution sources, with the air mass 45 originating from the southeast, along with biomass burning emissions (19%) from the 46 northeast, contributing significantly. Our research highlights that short-term, strict local 47 emission controls may not effectively reduce PM pollution, due to the non-linear 48 responses of  $PM_{2.5}$  to its precursors, further effective  $PM_{2.5}$  reduction requires a 49 comprehensive and extensive approach with a regionally coordinated and balanced 50 control strategy through joint regulation.

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#### 52 **1. Introduction**

53 China has implemented long-term clean air measures to cut down anthropogenic 54 emissions and improve air quality (Ge et al., 2020), resulting in a nationwide reduction 55 of average fine particulate matter (PM<sub>2.5</sub>, aerodynamic diameter  $\leq 2.5 \,\mu$ m) level from 50 μg m<sup>-3</sup> in 2015 to 30 μg m<sup>-3</sup> in 2020 (Zhou et al., 2022). However, this PM<sub>2.5</sub> 56 concentration remains significantly higher than the new World Health Organization 57 58 (WHO) guideline value of 5 µg m<sup>-3</sup> (WHO Global Air Quality Guidelines, 2021). Black 59 carbon (BC) is a ubiquitous component of PM2.5 that can mix with various species, and 60 the number fraction of BC-containing particles (BCc) can be higher than 50% of PM<sub>2.5</sub> 61 in China (Sun et al., 2022; Xie et al., 2020; Chen et al., 2020). Additionally, the 62 atmospheric aging of BCc involves complex chemical and physical processes, 63 influencing their mixing state, morphology, hygroscopic and optical properties, etc.,





ultimately impacting their climatic and health effects (Bond et al., 2013; Ramanathan
et al., 2008). Reducing the mass loading of BCc is therefore essential to comply with
the new WHO PM<sub>2.5</sub> guideline. Moreover, the insufficient understanding of complex
emission sources (e.g., fossil fuel and biomass burning), aging processes (e.g.,
coagulation, condensation, and cloud processing), and physical properties (e.g., mixing
state and coating composition) of BCc, hampering the effectiveness of air quality
remediation (Cappa et al., 2019; Kahnert, 2010; Sun et al., 2021).

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72 Yangzhou is located in the central region of Yangtze River Delta (YRD), at the junction 73 of the Yangtze River and, the Beijing-Hangzhou Grand Canal, which serves as a 74 prominent economic city, industrial-intensive area, and highly active inland shipping 75 node in East China. Due to the complex emissions and feedback with the East Asian 76 monsoons, this region is susceptible to anthropogenic aerosols, especially BCc particles 77 originating from chemical, steelmaking, coal-fired, petrochemical enterprises, and 78 transportation, etc. Extensive studies have investigated the responses of atmospheric 79 pollutants to emission changes during the COVID-19 lockdown measures in the YRD 80 (Chen et al., 2021; Li et al., 2020; Qin et al., 2021; K. Zhang et al., 2022). However, 81 the key chemical and physical processes specifically responsible for the BCc particles 82 in this region are still not fully understood. During the summer of 2021, Yangzhou 83 experienced a resurgence of COVID-19 with over 500 confirmed cases. In response, stringent public health measures were imposed from July 28th to September 10th, 84 85 including the closure of public transport, and suspension of non-essential industrial plants, restaurants, shopping malls, and entertainment clubs. People were also 86 87 mandated to quarantine at home. Unlike the nationwide COVID-19 lockdown in China 88 during the cold season of 2020 (Le et al., 2020; Sulaymon et al., 2021b), the summer 89 lockdown in Yangzhou was more localized but protracted, significantly altering local 90 anthropogenic emissions while neighboring cities maintained regular operations. This 91 scenario provides a unique opportunity to explore and compare the diverse mixing 92 states and aging process of BCc particles in different anthropogenic emission-intensive, 93 investigate the regional transportation of air pollutants in the YRD, enhance our 94 knowledge about the formation of BC-associated secondary components (Lei et al., 95 2021; Zhang et al., 2020) and understand emissions-meteorology interactions (Jiang et 96 al., 2021; Le et al., 2020) in the YRD.

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98 Studies on the effects of large-scale and short-term stringent emission control events on 99 air quality in China have been widely deployed, e.g., the 2008 Beijing Olympic Games 100 (Wang et al., 2010; Zhou et al., 2010), the 2015 Asia-Pacific Economic Cooperation 101 (APEC) (Zhu et al., 2015), the 2014 Nanjing Youth Olympic Games (Wang et al., 2022) 102 and the national COVID-19 lockdown in 2020 winter (Huang et al., 2021; Le et al., 103 2020; Li et al., 2020; Wang et al., 2020). Previous studies extensively investigated air 104 pollutant variations during the COVID-19 lockdown in the winter of 2020 across 105 different regions of the world. Stringent restrictions on industrial and vehicular





106 activities have resulted in significant reductions in gaseous pollutants and particulate 107 matter, not only in megacities (Chen et al., 2020; Jeong et al., 2022; Sun et al., 2020) 108 but also in middle-sized cities (Clemente et al., 2022; Wang et al., 2021; Xu et al., 2020) 109 and rural areas (Cui et al., 2021, 2020; Jain et al., 2021). Compared to the decreasing 110 trends observed in most cities worldwide, the level of PM<sub>2.5</sub> in Shanghai (Chang et al., 111 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 112 113  $PM_{2.5}$  comprising primary and secondary components. The reduction of primary 114 pollutants during lockdown resulted in a shift towards a higher proportion of secondary 115 aerosols, including inorganic and organic species, exhibiting a non-linear response to 116 emission changes (Zhang et al., 2021). Furthermore, some studies have suggested that 117 the increase in secondary aerosols during lockdown is due to the enhanced atmospheric 118 oxidative capacity resulting from the rise in ozone levels (Wang et al., 2021), 119 unfavorable meteorological conditions (Chien et al., 2022; Sulaymon et al., 2021a), 120 changes of local and regional emission sources (Feng et al., 2022), etc. However, most 121 previous studies focused on lockdown events during the cold seasons, studies on 122 summer lockdown events in China were very limited.

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124 To better understand the chemical compositions and aging characteristics of airborne 125 BCc particles in the YRD, we conducted ground measurements, spaceborne observations, and mass spectrometric analysis during the COVID-19 2021 summer 126 lockdown in Yangzhou. Besides, We employed potential source contribution function 127 128 (PSCF) analysis and a novel approach for distinguishing local sources to study the air 129 pollution regional transport in the YRD. This study investigated the impact of small-130 scale and short-term stringent emission controls on local ambient aerosol and the 131 mixing state of BCc particles, providing valuable insights for future air pollution control 132 measures.

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## 134 **2. Methods**

## 135 2.1 Sampling site and instruments

136 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 137 138 Monitoring Center (32.41°N, 119.40°E), Yangzhou, China (Figure 1). This sampling site is a typical urban site surrounded by residential areas, arterial roads, parks, 139 140 restaurants, and shopping centers. In this study, the measurement period was divided into three phases: the before-lockdown period (BLD: 30 June to 27 July 2021), the 141 142 lockdown period (LD: 28 July to 9 September 2021), and the after-lockdown period 143 (ALD: 10 September to 7 October 2021) (Figure 2). 144

145 A single-particle aerosol mass spectrometer (SPA-MS, Hexin Analytical Instrument Co.,

146 Ltd., China) was deployed during the field campaign to obtain chemical composition,





147 size distribution, and mixing state of individual PM2.5 particles. A PM2.5 cyclone (Model 148 URG-2000-30ED) and a Nafion dryer are equipped in front of the sampling inlet. 149 Individual particles are introduced into the SPA-MS through a critical orifice with a 150 flow rate of 3 L min<sup>-1</sup>. The vacuum aerodynamic diameters ( $D_{va}$ ) are determined using 151 the velocities derived from two continuous laser beams (diode Nd: YAG, 532 nm) 152 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 153 154 a Z-shaped bipolar time-of-flight mass spectrometer. A more detailed description of 155 SPA-MS can be found in previous studies (Li et al., 2011, Zhang et al., 2022). 156 157 PM<sub>2.5</sub> mass concentration was measured by a particulate matter monitor (XHPM2000E, 158 Xianhe, China). Nitrogen oxides ( $NO_x = NO + NO_2$ ), SO<sub>2</sub>, and ozone (O<sub>3</sub>) 159 concentrations were detected with a set of Thermo Fisher Scientific instruments (Models 42i, 43i, and 49i). The concentrations of 103 volatile organic compounds 160 161 (VOCs) in ambient air, comprising 57 ozone precursors (PAMS), 12 aldehydes and ketones, and 34 toxic organics (TO15), were continuously monitored at hourly intervals 162 using an online device (TH-300B, Tianhong, China). Meteorological parameters, 163

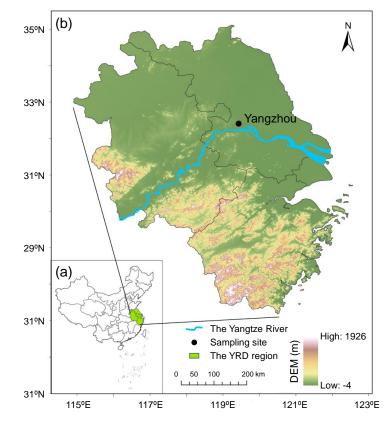
including ambient temperature (T), relative humidity (RH), precipitation (PCP), wind
 direction (WD), and wind speed (WS) were observed synchronously using an automatic

166 weather instrument (WXT530, Vaisala, Finland). All online data presented in this paper

167 were hourly averaged at local time (Beijing time, UTC+8).







#### 168

Figure 1. Location of (a) the Yangtze River Delta (YRD) in China and (b) the samplingsite in Yangzhou (Maps were generated by using Arcgis Pro).

## 171 2.2 Data analysis

#### 172 2.2.1 Satellite Product

173 Remote sensing of nitrogen dioxide (NO<sub>2</sub>) and sulfur dioxide (SO<sub>2</sub>) using satellite has 174 become a crucial tool for studying air pollution on a large spatial scale. In this study, 175 we utilized the Level 3 Near Real-Time Product of NO<sub>2</sub> (NRTI/L3 NO2) obtained from 176 the TROPOspheric Monitoring Instrument (TROPOMI) with a spatial resolution of 3.5×7 km<sup>2</sup> to analyze the distribution of total vertical column of NO<sub>2</sub> (Cooper et al., 177 2022). To avoid the obvious noises present in the NRTI/L3 SO2 data over clean regions, 178 179 we employed the SO2SMASS band from the Modern-Era Retrospective Analysis for 180 Research and Applications, version 2 (MERRA-2 SO2SMASS) with a spatial resolution of 69×55 km<sup>2</sup> to represent the distribution of SO<sub>2</sub> surface mass concentration 181 (Ukhov et al., 2020). We calculated and plotted the averaged 2-dimensional data of 182 183 NRTI/L3 NO2 and SO2SMASS during BLD and LD over the region of interest 184 (17.93~54.74 °N, 71.21~142.23 °E) using Google Earth Engine (Gorelick et al., 2017). 185 The integration of MERRA-2 and TROPOMI measurements has provided a more 186 comprehensive understanding of the sources and distributions of NO2 and SO2





187 facilitating the evaluation of the impact of human activities on air quality.

#### 188 2.2.2 Geographic Source Analysis

189 The potential source contribution function (PSCF) analysis, based on the Hybrid 190 Single-Particle Lagrangian Integrated Trajectory (HYSPLIT) model, can be employed 191 to identify regional sources of air pollutants. Before conducting the PSCF analysis, 36 192 hours of air mass backward trajectories with one-hour resolution at 500 m above ground 193 level were calculated using the wind data from the Global Data Assimilation System 194 (GDAS) provided by the National Oceanic and Atmospheric Administration (NOAA) 195 (Wang et al., 2009). An open-source software MeteoInfo (Wang et al., 2014) was 196 utilized for the PSCF analysis. The whole study area (110.1~133.4 °E and 21.3~39.9 197 <sup>o</sup>N) covered by the trajectories was divided into thousands of cells with a spatial 198 resolution of  $0.1^{\circ} \times 0.1^{\circ}$ . The PSCF was simulated according to the following equation:

$$PSCF_{ij} = \frac{m_{ij}}{n_{ij}} \tag{1}$$

200 where  $PSCF_{ii}$  is the conditional probability that the grid cell (i, j) was a source of the 201 species found in high concentration (Hopke et al., 1993);  $n_{ij}$  is the number of all 202 trajectories passing through this grid cell, and  $m_{ij}$  is the number of trajectories. In this 203 study, the pollution criterion values for different BCc particle types were set as the 75<sup>th</sup> 204 percentile of hourly average number fractions, respectively. To further improve the accuracy of the PSCF analysis and minimize analytical uncertainties, the Weighted 205 PSCF (WPSCF) functions as shown in Equation (2~3) were applied (Polissar et al., 206 207 1999). The weight  $(W_{ij})$  for each grid cell was determined based on the number of 208 trajectory endpoints  $(n_{ii})$  as follows:

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

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

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

#### 212 2.2.3 SPA-MS Data Analysis

213 In total, 1649574 particles were analyzed during the entire observation period. The size 214 and chemical composition of single particles were analyzed using the Computational Continuation Core (COCO V1.4) toolkit in MATLAB 2022 (The MathWorks, Inc.). 215 216 Our focus was on BCc particles, which were 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., 217 218 2021). An adaptive resonance theory-based neural network algorithm (ART-2a) was 219 applied to classify the measured individual particles based on the presence and intensity 220 of ion peaks, with a vigilance factor of 0.75, a learning rate of 0.05, and 20 iterations 221 (Song et al., 1999).





# 222 3. Results and discussion

# 223 3.1 Overview of field observations

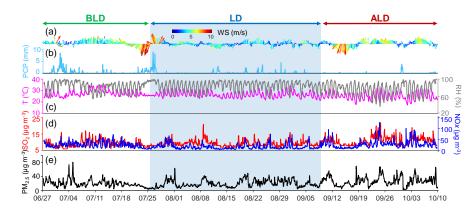
224 Figure 2 presents the temporal variations of meteorological parameters, PM2.5, NOx, and SO<sub>2</sub> concentrations during the entire observation. During the BLD stage, the mean 225 226 temperature (T) was 28.2±2.6 °C, with an average relative humidity (RH) of 81.4± 11.1%. The prevailing winds originated from the south and southeast, with a mean wind 227 228 speed (WS) of  $3.4\pm0.9$  m s<sup>-1</sup>. Notably, PM<sub>2.5</sub>, NO<sub>x</sub>, and SO<sub>2</sub> were dramatically reduced 229 at the end of the BLD period due to a high precipitation event, and the data collected during the precipitation were excluded from the analysis. In comparison, the LD period 230 231 saw a decline in temperature to  $26.2\pm2.4$  °C, a reduction in WS to  $2.3\pm0.8$  m s<sup>-1</sup>, and 232 an increase in RH to 86.6±10.1%. Additionally, Figures S2b~c exhibit uniform 233 distributions of RH and boundary-layer height (BLH) across the YRD during the LD period. The resemblance of meteorological elements with other cities in the YRD (Qian 234 235 et al., 2022; Wang et al., 2022) and the effective removal of the pollutants accumulated during the BLD stage imply that Yangzhou is mainly affected by upwind transmission 236 during the LD period, providing favorable conditions for investigating the regional 237 transport of BCc particles in the YRD during summer. Subsequently, the temperature 238 declined to 25.2±3.5 °C, the WS increased to 3.2±1.4 m s<sup>-1</sup>, and RH decreased to a 239 240 lower level of  $74.7 \pm 15.0\%$  during the ALD period.

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Further, surface concentrations of NO<sub>x</sub> (18.9  $\mu$ g m<sup>-3</sup>) and TVOC (55.0  $\mu$ g m<sup>-3</sup>) were the 242 243 lowest during the LD period compared to those of the BLD and ALD periods, whereas 244 the surface O<sub>3</sub> concentration showed an increase of 12.6  $\mu$ g m<sup>-3</sup> (19%) during the LD 245 period compare to the BLD period, which may attribute to the reduction of fresh NO 246 emissions that alleviates O3 titration (Steinfeld, 1998). However, the average concentrations of PM2.5 (19.9 vs. 21.2 µg m<sup>-3</sup>) and SO2 (9.4 vs. 9.5 µg m<sup>-3</sup>) were very 247 248 close between BLD and LD stages (Figure 3). Following the end of lockdown, social 249 activities gradually resume in Yangzhou City, leading to an apparent increase in all 250 observed pollutants during the ALD period. For instance, the relative increases from LD to ALD were 71% for NO<sub>x</sub>, 22% for SO<sub>2</sub>, 55% for TVOC, 30% for O<sub>3</sub>, and 29% for 251 252 PM<sub>2.5</sub> (Figure 3), respectively.



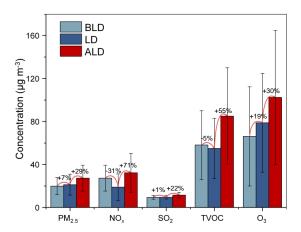




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Figure 2. Temporal variations of (a) wind direction (WD) and wind speed (WS), (b)

255 precipitation (PCP), (c) temperature (T) and relative humidity (RH), (d) concentrations 256 of NO<sub>x</sub> and SO<sub>2</sub>, and (e) mass loading of  $PM_{2.5}$ .



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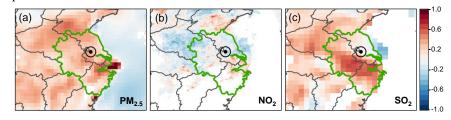
Figure 3. Ground-based observations of  $PM_{2.5}$ ,  $NO_x$ ,  $SO_2$ ,  $O_3$ , and TVOC concentrations in Yangzhou. The figure compares the averages during the BLD (bluegrey), LD (dark-blue), and ALD (crimson) periods. Error bars indicate SDs over different lockdown periods.

262 In addition to ground measurements, satellite retrieved PM<sub>2.5</sub>, NO<sub>2</sub>, and SO<sub>2</sub> data over the entire region of eastern China were also investigated. Results show that the hotspots 263 264 of those pollutants were predominantly located over eastern China, e.g., the YRD region, during both the BLD and LD periods (Figure 3). Figure 4 displays the regional 265 fractional changes in mean PM<sub>2.5</sub>, NO<sub>2</sub>, and SO<sub>2</sub> levels from the BLD to LD periods in 266 the YRD, indicating a 29%, 25%, and 23% increase, respectively. In comparison, 267 Yangzhou city experienced lower increases in these air pollutants, with changes of 25%, 268 -23.8%, and 2.9% for PM<sub>2.5</sub>, NO<sub>2</sub>, and SO<sub>2</sub>, respectively (remarkable NO<sub>2</sub> decrease). 269 270 Such results highlight the short-term, limited-scale, and human-induced reduction in air 271 pollutants as a result of the lockdown measures in Yangzhou, and demonstrate the





effectiveness of regional stringent emission control in reducing local atmosphericpollutant concentrations.



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Figure 4. The fractional changes (i.e.,  $100 \times (LD - BLD)/BLD$ ) of (a) PM<sub>2.5</sub>, (b) NO<sub>2</sub>, and (c) SO<sub>2</sub> between BLD and LD periods based on spaceborne measurement. Calculations were only conducted for the regions with PM<sub>2.5</sub> > 10 µg m<sup>-3</sup>, NO<sub>2</sub> > 0.2 Dobson units (DU), and SO<sub>2</sub> > 0.2 DU in the BLD period. The circle symbols in the maps indicate the location of Yangzhou, and the green region represents the YRD (1 DU = 0.4462 mmol m<sup>-2</sup>).

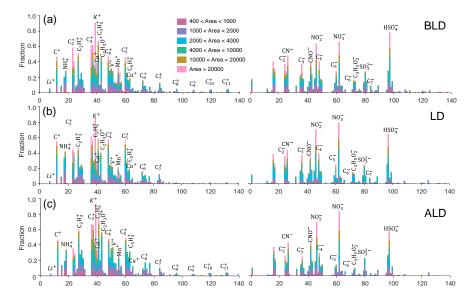
# 281 3.2 Chemical composition and size distribution of individual

# 282 BCc particles

283 Based on the SPA-MS analysis, a total of 1068362 BCc particles were collected over 284 the whole study period. The BCc particles accounted for 58.8%, 67.7%, and 56.5% of the total number of measured particles in the BLD, LD, and ALD periods, respectively. 285 286 Figure 5 shows the normalized average mass spectra of BCc particles during three 287 periods. Ion height in each spectrum reflects the number fraction of the detected BCc particles with the corresponding ion to the total BCc particles, while colors represent 288 289 peak area ranges of detected ions. As shown in Figure 5, BCc particles in BLD, LD, 290 and ALD shown similar mass spectra at m/z < 100, with common peaks including carbon ion clusters ( $C_n^{\pm}$ ,  $n = 1 \sim 7$ ),  $27[C_2H_3]^+$ ,  $37[C_3H]^+$ ,  $43[C_2H_3O]^+$ ,  $51[C_4H_3]^+$ , 291  $63[C_5H_3]^+$ ,  $46[NO_2]^-$ ,  $62[NO_3]^-$ , and  $97[HSO_4]^-$ . However, the abundance of large m/z292 293 carbon ions ( $C_n^{\pm}$ , n > 7) in both BLD and ALD periods was approximately 1.5 times that 294 in the LD. The result was a clear reflection of less local vehicle emissions during the LD period (Liu et al., 2019), in line with aromatics, e.g.,  $119[C_9H_{11}]^+$ . 295







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

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301 BCc particles were further classified into 12 types based on the differences of chemical 302 features and temporal variations as shown in Table S1. Fresh BC particles (BC-fresh) 303 are those freshly emitted without undergoing significant atmospheric processing (Ding et al., 2021). Five types of BC-fresh particles were identified according to the 304 305 characteristics ion markers: (i) BC-pure is dominated by carbon clusters  $(C_n^{\pm})$  with minor ion signals of secondary inorganic species, such as 46[NO<sub>2</sub>]<sup>-</sup>and 97[HSO<sub>4</sub>]<sup>-</sup> 306 307 from nitrate and sulfate, respectively (Xie et al., 2020); (ii) BCc particles from biomass 308 burning (BB) are characterized by ion signals at m/z 39[K]<sup>+</sup>, 45[CHO<sub>2</sub>]<sup>-</sup>, 59[C<sub>2</sub>H<sub>3</sub>O<sub>2</sub>]<sup>-</sup>, 309 and  $73[C_3H_5O_2]^-$ , with relative peak area (RPA) more than 0.5 (Silva et al., 1999); (iii) 310 coal combustion BCc particles (CC) typically include small carbon clusters ( $C_n^{\pm}$ , n = 311 1~4), metal elements (e.g., 7[Li]<sup>+</sup>, 23[Na]<sup>+</sup>, 27[Al]<sup>+</sup>, 56[Fe]<sup>+</sup>, 63[Cu]<sup>+</sup> and 312  $206/207/208[Pb]^+$ ), and organic carbon  $(38[C_3H_2]^+, 43[C_2H_3O]^+)$  peaks in the positive 313 mass spectrum, while the strong signals of secondary inorganic species (46[NO<sub>2</sub>]<sup>-</sup>, 314 43[AlO]<sup>-</sup>, 62[NO<sub>3</sub>]<sup>-</sup>, 80[SO<sub>3</sub>]<sup>-</sup>, 97[HSO<sub>4</sub>]<sup>-</sup>) in the negative ion mode suggest that CC 315 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 316 317 of ion signals at *m/z* of 40[Ca]<sup>+</sup>, 51[V]<sup>+</sup>, 55[Mn]<sup>+</sup>, 67[VO]<sup>+</sup>, 46[NO<sub>2</sub>]<sup>-</sup>, 62[NO<sub>3</sub>]<sup>-</sup>, and 318  $79[PO_3]^-$ , as well as high loadings of organic carbon  $(41[C_3H_5]^+, 43[C_2H_3O]^+)$  and 319 carbon clusters ( $C_n^{\pm}$ , n = 1~4) ion peaks (Yang et al., 2017); (v) BCc particles that are 320 internally mixed with more than one type (BB, CC, and VE) are categorized as Mix 321 type (Sun et al., 2022).





323 Aged BC particles, denote as BC-aged, undergo a series of chemical reactions and 324 physical transformations. These processes typically lead to changes in their morphology, 325 hygroscopicity, and optical properties as they are coated with other materials (He et al., 326 2015). Six types of BCc particles are classified as BC-aged and are further grouped into 327 BCOC and BC-SNA, depending on whether they contain mainly organic carbon (OC) 328 or sulfate/nitrate/ammonium (SNA). First, BCOC types indicate BC-aged particles that internally mixed with OC. These particles are characterized by the presence of carbon 329 330 clusters  $(C_n^{\pm})$  and  $C_n H_m^{+}$  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-331 332 SNA indicating the mix with secondary inorganic species. Additionally, BCOC 333 particles with negative mass spectra dominated by nitrate ions (46[NO<sub>2</sub>]<sup>-</sup> and 62[NO<sub>3</sub>]<sup>-</sup>) 334 or sulfate ions (97[HSO4]<sup>-</sup>) are referred to as BCOC-N or BCOC-S, respectively; 335 otherwise, BCOC particles showing similar peak areas of nitrate and sulfate are named BCOC-SN. Furthermore, The BC-SNA particles are further categorized as BC-N, BC-336 337 S, and BC-SN based on similar principles. Note the remaining particles that cannot be classified into neither BC-fresh or BC-aged ones are denoted as BC-other. More details 338 339 of BCc particle types are shown in Table S1 and Figure S1 in the Supplement.

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341 During the BLD period, the average number fraction of BC-fresh particles was 36% 342 with sizes mainly concentrated at ~500 nm, while the mode size of BC-aged particles was ~520 nm (Figure 7). The predominant BCc types during the BLD period were 343 BCOC-S and BC-S (24% and 12% by number), likely because sulfate was removed 344 less efficiently than organic matter (OM) and NO<sub>3</sub> by heavy precipitation, especially 345 346 during the warm seasons (Isok äänt äet al., 2022). As shown in Figure 7c and d, the peak 347 size of BC-SNA was larger than that of BCOC in all periods, indicating that organics 348 coated BCc generally had a relatively thin coating compared to those coated by 349 secondary inorganic species, which is consistent with previous studies (Sun et al., 2016; 350 Wang et al., 2019).

351

During the transition of BLD period ( $PM_{2.5}$ : 19.9 µg m<sup>-3</sup>, O<sub>3</sub>: 66.2 µg m<sup>-3</sup>, NO<sub>x</sub>: 27.3 µg 352 m<sup>-3</sup>) to LD period (PM<sub>2.5</sub>: 21.2 µg m<sup>-3</sup>, O<sub>3</sub>: 78.8 µg m<sup>-3</sup>, NO<sub>x</sub>: 18.9 µg m<sup>-3</sup>), heavy 353 precipitation occurred on July 28<sup>th</sup> (the day before lockdown) and scavenged a majority 354 of the pollutants (PM<sub>2.5</sub>: 4 µg m<sup>-3</sup>, O<sub>3</sub>: 35 µg m<sup>-3</sup>, NO<sub>x</sub>: 8 µg m<sup>-3</sup>). After that, the strict 355 356 lockdown measures were carried on and the primary emissions were abruptly cut down. 357 As a result, the number fraction of BC-fresh particles significantly decreased from 37% 358 to 28% and that of VE-type particles dropped from 12% to 3% (by number). As shown 359 in Figure 3, with the decrease in NOx, an obvious enhancement of O3 was observed during the LD period. According to previous studies (Huang et al., 2021; Laughner et 360 361 al., 2021), large reduction of  $NO_x$  could promote the formation of  $O_3$  under a VOC-362 limited regime and enhance the oxidation capacity of the local atmosphere, which made 363 the number fraction of BC-aged particles increased from 64% in the BLD to 72% in the 364 LD period (Figure 6a), indicating the lockdown measures could accelerate aging of BCc





365particles through complicated chemical reactions and/or physical coagulation. We also366found the most abundant type of BCc particles changed from BCOC-S (24% by number)367in the BLD to BC-N (25%) in the LD (Figure 6a). Furthermore, despite the abrupt368reductions of NOx due to city lockdown, it should be aware that the PM2.5 concentration369slightly increased during the LD period, highlighting the non-linear relationship370between primary emissions and PM2.5 levels.

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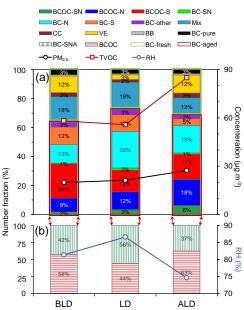
During the ALD period (PM<sub>2.5</sub>: 25.9 µg m<sup>-3</sup>, NO<sub>x</sub>: 27.9 µg m<sup>-3</sup>, TVOC: 76.4 µg m<sup>-3</sup>), 372 the number fraction of BC-fresh particles increased from 28% to 31%, and the fraction 373 374 of VE particles also increased from 3% to 12% (by number) (Figure 6a). Notably, the 375 size distributions of BC-fresh and BC-aged particles presented relatively small peaks 376 at 690 nm and 820 nm during the ALD period, in addition to the prominent peaks at 377 490 nm and 500 nm, which were different from those in the BLD and LD periods. These 378 small peaks were relatively close to the dominant sizes of BC-fresh and BC-aged 379 particles during the LD period (Figure 7). This result suggests that a substantial number 380 of BCc particles with small sizes (around 500 nm) after the lockdown was lifted in 381 Yangzhou, owing to the sudden enhancement of primary emissions; on the other hand, 382 particles with large diameters (>690 nm) may have formed due to the participation of 383 more trace reactive gases (e.g., NO<sub>x</sub>, SO<sub>2</sub>, and VOCs) in continuous aging reactions, 384 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 385 periods. This hypothesis was also supported by the increased number fraction of 386 BCOC-SN during the ALD period (Figure 6a). Similar findings have been reported in 387 388 the North China Plain (NCP) and the YRD during cold seasons, where thicker coatings 389 on secondary aerosols were also observed under lower RH (<70%) (Zhang et al., 2021). 390 This might be due to that particles with more organics and nitrate can result in earlier 391 deliquescence and provide aqueous surfaces that facilitates heterogeneous formation of 392 secondary species under relatively low RH (Zhang et al., 2021). Among the three 393 periods, the difference between the mode sizes of BC-aged and BC-fresh particles was 394 the smallest (10 nm) during the ALD period (Figure 7a and b). This size reduction can 395 be attributed to the increased BCOC and hydrophobic primary particles after lockdown 396 (Figure 6). Because the internally mixed BCOC and hydrophobic primary particles may 397 constrain further growth of secondary BC-SNA particles (Liu et al., 2016; Zhang et al., 398 2018), thereby leading to smaller-sized BC-aged particles. Moreover, the differences of 399 BCc mode sizes between ALD and BLD periods also reveals an interesting fact that the 400 lockdown effect may not only affect air quality during lockdown, but also can influence 401 the air quality even after lockdown, as the resumed emissions after lockdown may be 402 subjected to different chemistry from that before lockdown. 403

404 Throughout the entire observation, the changes in the number fraction of BC-SNA
405 showed consistency with the variations in RH (Figure 6b), indicating that BC tends to
406 mix with ammonium sulfate and ammonium nitrate under high RH conditions overall.





407 Meanwhile, the number fraction of BCOC had similar patterns of change as TVOC, 408 suggesting that high TVOC levels may facilitate the coating of organics on BC cores. 409 Figure 8 displays the number fraction of BCc species as a function of PM<sub>2.5</sub>. Overall, 410 as PM2.5 levels increased, the number fraction of BC-aged particles also increased, 411 while the proportion of BC-fresh particles decreased during the BLD and LD periods, 412 indicating a clear transition from BC-fresh particles to more aged ones. However, the increase in PM2.5 was driven by BCOC-S during the BLD period (Figure 8a), whereas 413 414 BC-N played a vital role in the PM<sub>2.5</sub> increase during the LD period (Figure 8b). Interestingly, the concentration of NO<sub>x</sub>, the primary precursor of BC-N, decreased by 415 416 31% and 41% during the LD period compared to the BLD and ALD periods, 417 respectively (Figure 3). Despite the significant decrease, the proportion of BC-N during 418 the LD period was unexpectedly higher than those during the BLD and ALD periods, 419 indicating a strong non-linear response of nitrate in BCc particles to NO<sub>x</sub>, very likely 420 due to much faster conversion of NOx to nitrate upon enhanced atmospheric oxidation 421 capacity; additionally, the high proportion of BC-N during the LD period might be 422 attributed to regional transport, similar to that in Shanghai during 2020 winter lockdown 423 (Chang et al., 2020).



424

425 Figure 6. Number fractions of BCc particles. (a) The number fractions of different BCc

426 particles along with the concentrations of  $PM_{2.5}$  and total volatile organic compounds

- 427 (TVOC). (b) The number fractions of different types of BC-aged particles along with
- 428 relative humidity (RH).





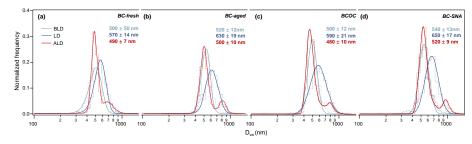
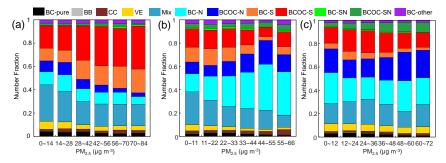




Figure 7. Size distribution of different types of BCc particles during different lockdown
in Yangzhou. (a) BC-fresh particles, (b) BC-aged particles, (c) BCOC particles, and (d)
BC-SNA particles. The Log-normal distribution was used to fit the unimodal size
distribution, and the Lorentz distribution was used to fit the bimodal size distribution.
The corresponding mode sizes (with the standard deviations) are also shown.

435



436

Figure 8. Variations of number fractions of BCc particle types with PM<sub>2.5</sub> mass concentrations during (a) the BLD period, (b) the LD period, and (c) the ALD period.

# 439 3.3 Chemical aging of BCc Particles

440 As shown in Figure 5, in the average positive mass spectra of total BCc particles, the 441 peak areas of  $C_n^+$ , OM, and metals contributed to more than 95% of the total, while 442 nitrate and sulfate peak areas accounted for more than 90% of the negative mass 443 spectral signal. To better elucidate the aging processes of BCc particles during different lockdown periods, we summed the carbon clusters  $C_n^{\pm}$  (n = 1~5, accounting for more 444 445 than 99% of  $C_n$ ) peak areas to represent BC, and the total peak area of sulfate, nitrate, 446 and ammonium (SNA) to represent the second inorganic components coated on BC. 447 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 448 449 coating materials, except for metals, of BCc particles. The changes in the mixing state 450 and morphology of BCc particles can provide insights into their aging characteristics, 451 as reported previously (Kandler et al., 2018; Moffet et al., 2013). In this study, we use 452  $OC/C_n$  and  $SNA/C_n$  ratios to describe different types of chemical components coated 453 on BC-fresh, and we use the ratio of the mode size of BC-aged ( $D_{aged}$ ) to that of 454 contemporaneous BC-fresh (D<sub>fresh</sub>) to represent the aging degree of BCc particles. 455





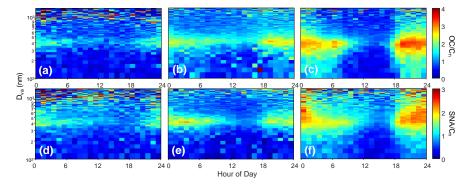
456 Figure 9 illustrates the diurnal variations of the  $OC/C_n$  and  $SNA/C_n$  ratios along with 457 the size distribution of BCc particles during different periods. Throughout the entire 458 observation, we observed that both  $OC/C_n$  and  $SNA/C_n$  increased during nighttime and 459 decreased during daytime. These variations showed the prominent enhancements of 460 nocturnal OM and SNA, which could be attributed to the accelerated gas-to-particle 461 partitioning and nocturnal secondary formation of organic/inorganic components under high relative humidity (RH > 85%) and relatively stagnant air (WS  $< 3 \text{ m s}^{-1}$ ) (Figure 462 463 S4). It is worth noting that from the BLD period to the LD and ALD periods, the 464 intensity of diurnal variations of OC/Cn and SNA/Cn increased obviously. This 465 discrepancy can attribute to several reasons. During the BLD period, the frequent 466 precipitations effectively scavenged the particles (Isok äänt ä et al., 2022). In contrast, 467 stronger solar radiation and higher ozone level during the LD period promoted 468 photochemical formations of OC and SNA; After lockdown, more precursors due to increased local emissions may lead to more production of secondary components than 469 470 that during the BLD period as explained earlier. These results indicate that the aging 471 process and mixing state of BCc particles depend strongly on weather conditions and 472 anthropogenic emission structures in urban cities.

473

474 As shown in Figure 9, BCc particles with ~400 nm Dva exhibited significant diurnal 475 fluctuations in the OC/Cn and SNA/Cn ratios, during the LD period. Moreover, there was a noticeable increase in the proportion of BC-SNA particles during nighttime when 476 477 RH was relatively high. These observations suggest that nighttime heterogeneous 478 hydrolysis may be considered as a key mechanism responsible for the formation of 479 BCOC and BC-SNA particles. According to Jacobson (2002), coagulation can be 480 significant between particles with sizes <100 nm and >1 µm but insignificant for particles of >300 nm, when the total particle number concentration is higher than 481 482 10<sup>4</sup> cm<sup>-3</sup>. During the LD period, the OC/C<sub>n</sub> and SNA/C<sub>n</sub> ratios of BCc particles with 483  $\sim$ 400 nm D<sub>va</sub> exhibited pronounced diurnal variations (Figure 9) and the number 484 fraction of BC-SNA increased obviously. Despite the difference between Dva and 485 physical diameter, such results imply that chemical reactions should be considered as 486 the major pathway for BCOC and BC-SNA particles of ~400 nm Dva, while the large-487 sized BC-aged particles (>1  $\mu$ m) may be partially from physical coagulation. Moreover, 488 the larger peak Dva (~600 nm) and higher Daged/Dfresh ratios (1.11) were observed 489 compared to those of the BLD (~510 nm, 1.03) and the ALD periods (~500 nm, 1.02) 490 (Figure 7). Since RH was significantly higher during LD period (average RH of 86.6%) 491 than the BLD period (average RH of 81.4%) and ALD period (average RH of 74.7%), 492 this result again supports that aqueous or heterogeneous reactions might play a more 493 important role to facilitate the chemical conversion of trace reactive gases (e.g., SO<sub>2</sub>, 494 NO<sub>x</sub>, and VOCs) and then formed a thicker coating on the surfaces of BC cores, leading 495 to evident growth in the size of BCc particles. In addition, this aqueous or 496 heterogeneous process during the LD period likely converted partially coated particles 497 to fully thickly coated BCc particles as well (Figure 11).







498

Figure 9. Diurnal variations of the ratios of OC/C<sub>n</sub> and SNA/C<sub>n</sub> with a size distribution
of BCc particles during (a, d) the BLD period, (b, e) the LD period, and (c, f) the ALD
period.

# 502 3.4 Source of BCc particles during lockdown

503 In addition to local emissions, regional transport plays a significant role in influencing 504 the pollutant levels. Due to the emergent lockdown in Yangzhou, local emissions were 505 strictly limited, while surrounding cities were still as usual, it is therefore interesting to 506 investigate source areas of BCc sources under such scenario. Besides, the air pollutants 507 were significantly influenced by regional transport, presenting an ideal opportunity to 508 investigate the transmission and source characteristics of BCc particles in the YRD 509 during summer. Here, the PSCF method was used to qualitatively simulate the source 510 probability distributions of the specific BCc particle types (BC-fresh, BC-aged, BCOC, 511 and BC-SNA) during the LD period. The results of the potential source regions and 512 clustering analysis are presented in Figure 10.

513

514 As shown in Figure 10, the hotspots of potential sources for the four particle types 515 exhibited strong agreements with each other and primarily concentrated in the southeast 516 of Yangzhou, especially along the coast of the Yangtze River, with the WPSCF greater 517 than 0.6. These hotspot areas also encompassed chemical enterprises, power plants, 518 petrochemical industrial parks, and the Yangtze River Ship Channel in the YRD. 519 Moreover, BCc particles and gaseous emissions from the YRD city cluster, heavy 520 industries, and ship diesel engines, can easily impact the air quality of surrounding 521 downwind regions. This evidence suggests that the region of southeast Yangzhou and 522 lower reaches of Yangtze River are major source areas for the regionally transported 523 BCc particles in Yangzhou during lockdown.





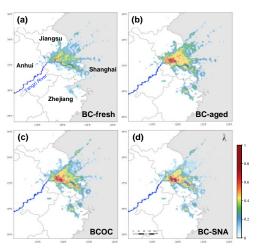


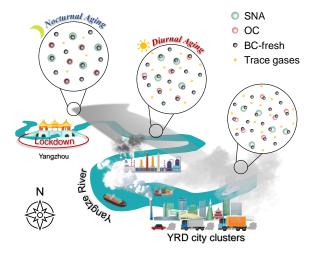


Figure 10. The potential sources areas of different BCc particles during the LD period.
(a) BC-fresh. (b) BC-aged. (c) BCOC. (d) BC-SNA.

According to Luo et al. (2023), regional transport of pollutants can occur near surface 527 from upwind areas when the wind speed (WS) exceeds 2 m s<sup>-1</sup>. Figure S4b shows that 528 the mean daytime WS was 3 m s<sup>-1</sup>, indicating that both BC-fresh and BC-aged particles, 529 530 along with trace gases (e.g., SO<sub>2</sub>, NO<sub>x</sub>, and VOCs), originating from the hotspot areas, could be transported effectively to Yangzhou. Additionally, the average size of BCc 531 532 particles remained around 0.6 µm at daytime (Figure S4c), suggesting that BCc particles could undergo continual aging reactions under relative lower RH, but 533 producing relatively thinly coated BCc particles with smaller sizes than those at 534 nighttime (average size of 0.65 µm) (Figure 11). The mean nocturnal WS decreased to 535 536 2 m s<sup>-1</sup>, indicating that the regional atmosphere becomes stagnant (Figures S4a, b). As mentioned earlier and underscored here again, this stagnant and humid atmospheric 537 538 condition may promote aqueous or heterogeneous reactions, likely further leading to the production of more thickly coated BCc particles than daytime ones (Figure 11). 539







540

541 **Figure 11.** A schematic diagram of the transportation of air pollutants and ageing 542 process from the YRD city cluster to Yangzhou during the 2021 summer COVID-19

543 lockdown.

# 544 3.5 Local and non-local source analysis

Since there was a heavy precipitation on July 28<sup>th</sup> (the day before lockdown) which 545 546 removed most atmospheric pollutants, the air pollutants might be influenced mostly by 547 regional transport as local emissions were significantly cut down during the LD period. 548 As a comparison, the pollution during the ALD period might be caused by both local 549 emissions and regional transport. Additionally, the meteorological conditions were 550 relatively stable during the LD and ALD periods (Figure 2a~c); the trajectories of these 551 two periods were both categorized into similar 3 clusters, indicating stable regional 552 transport of the pollutants from the southeast, southwest, and northeast (Figure 12). The 553 lockdown event with favorable meteorological conditions provided a valuable 554 opportunity to investigate emissions-meteorology interactions in YRD during summer. 555 Here, we propose a method to roughly estimate the local and non-local proportions of 556 for each type of BCc particles in Yangzhou during the ALD period (representing the 557 usual emission condition)

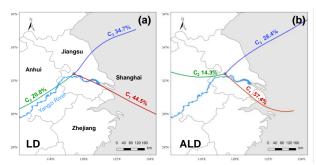
558 
$$[PM]_{i,i}^{non-local} = [PM]_{i,i}^{LD} \times t_i^{ALD}/t_i^{LD};$$
(4)

559 
$$[PM]_{i,i}^{local} = [PM]_{i,i}^{ALD} - [PM]_{i,i}^{non-local}.$$
 (5)

For Equation (4~5), the duration of the  $i^{th}$  cluster in different periods is represented by t<sup>LD</sup><sub>i</sub> and t<sup>ALD</sup><sub>i</sub>. The sum of the hourly number density of the  $j^{th}$  type of particulate matter in the  $i^{th}$  cluster during the LD period is denoted as  $[PM]^{LD}_{i,j}$ . Similarly,  $[PM]^{non-local}_{i,j}$ and  $[PM]^{local}_{i,j}$  indicate the summed hourly number density of specific types of BCc particles from non-local and local sources in the  $i^{th}$  cluster during the ALD period, respectively.

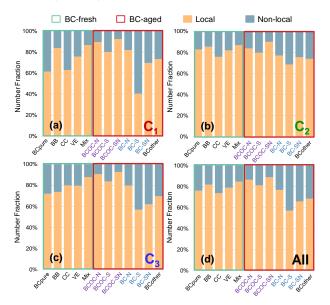






566

**Figure 12.** Back-trajectory analysis during **(a)** LD and **(b)** ALD period. The corresponding percentages of total trajectories for Cluster1 (C<sub>1</sub>, red), Cluster2 (C<sub>2</sub>, green) and Cluster3 (C<sub>3</sub>, blue) are also shown.



570

Figure 13. Number fraction of local and non-local in different types of particles in (a)
Cluster1 (C<sub>1</sub>), (b) Cluster2 (C<sub>2</sub>), (c) Cluster3 (C<sub>3</sub>), and (d) all clusters during the ALD
period. The purple labels represent BCOC particles and the blue labels represent BCSNA particles.

575

576 By using this method, analysis results of the sources of all types of BCc particles from 577 different clusters are presented in Figure 13a $\sim$ c. Cluster1 (C<sub>1</sub>), originating from the 578 economically developed southeast region, accounted for 57.4% of total trajectories 579 during the ALD period. C<sub>1</sub> exhibited the highest proportions of non-local fresh BCc 580 (BC-pure, 38.6%) and coal combustion BCc (CC 37.1%), along with vehicular ones 581 (VE 24.3%), underscoring the substantial impact of heavy industry and transportation 582 from the southeastern YRD during summer (Figure 13b). Notably, non-local 583 contribution of BC-S was dominant in C1, indicating significant regionally transported 584 sulfate. Cluster2 (C2), originating from the southwest region covering Nanjing and





585 eastern Anhui province, accounted for 14.3% of all trajectories (Figure 12b). The non-586 local proportions for all particle types were around 20%, indicating that local emission 587 was dominant (Figure 13b). Cluster 3 (C<sub>3</sub>), originating from the East China Sea and 588 passing through the vast cultivated area in northeastern Jiangsu Province, contributed 589 28.4% of the total trajectories (Figure 12b). A relatively high proportion of BCc 590 particles generated by non-local biomass-burning emissions (BB, 26.4%) was observed in C<sub>3</sub>, indicating a correlation between BCc particles from the northeastern YRD and 591 open-field burning of agricultural residues during summer (Figure 13c). 592

593

594 Regarding the number fraction of local and non-local contributions for the whole 595 campaign (Figure 13d), the proportion of local BC-N particles (~80%) exceeded that 596 of BC-S (~60%; note non-local contribution of BC-S even dominated over local 597 counterpart in C<sub>1</sub>), suggesting that sulfate-associated BCc particles were more likely from regional transport than those of nitrate-associated ones. The proportion of non-598 599 local BC-aged particles was relatively higher than the BC-fresh particles naturally, as BC-aged particles intercepted more secondary species during the regional transport 600 601 than freshly emitted BCc particles. Furthermore, the proportion of local BCOC particles 602 exceeded that of BC-SNA particles, implying a strong relationship between BCOC 603 particles and local emission, whereas more BC-SNA particles were likely associated 604 with regional transport. Overall, BCc particles was predominantly local (78%) in 605 Yangzhou during normal summertime. However, BCc particle from coal combustion 606 (CC, 26%) and vehicle emission (VE, 21%) transported from the southeast, as well as 607 biomass burning-related emissions (BB, 19%) from the northeast, were also significant 608 contributors that should not be ignored (Figure 13d). These findings highlight the 609 importance of considering both local and regional sources, as well as understanding the 610 transport characteristics of different types of BCc particles for air quality management.

# 611 **4. Conclusions and implications**

During the summer of 2021, the COVID-19 lockdown imposed in Yangzhou resulted 612 613 in a significant decrease in anthropogenic emissions from traffic and manufacturing 614 sectors. To examine the effects of this lockdown, we utilized spaceborne observations, 615 ground-based measurements, and particularly SPA-MS analysis to explore the 616 variations, aging characteristics, and sources of BCc particles in the YRD. We showed 617 that the strict emission controls effectively reduced local gaseous pollutants. However, 618 the decline in NO<sub>x</sub> (-30.1%) and TVOC (-5.3%) levels might on the other hand result 619 in increased ozone (+19.0%), leading to a rise in BC-aged particles and a slight 620 elevation in PM2.5 levels during the lockdown. Our results revealed a strong non-linear 621 response of PM<sub>2.5</sub> and O<sub>3</sub> to the gaseous precursors. 622

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-





626 aged particles (73%) during the lockdown due to enhanced oxidizing capacity and high 627 relative humidity (RH > 85%). The BC-fresh particles tended to mix with SNA under 628 high RH conditions, while high TVOC levels were accompanied by BCOC formation. 629 However, BCOC particles generally exhibited smaller sizes compared to BC-SNA 630 particles. Moreover, we postulate that aqueous or heterogeneous reactions might be 631 important to generate BCOC and BC-SNA particles, especially ones with ~400 nm Dva, while coagulation might play a more prominent role in larger BC-aged particles. The 632 633 aging process during the LD period promoted the conversion of partly coated particles 634 to totally coated ones, with larger diameters (~600 nm) and thicker coatings. 635 636 Furthermore, our study highlights that local emissions were the main source of BCc 637 particles in Yangzhou during normal summertime. However, regional transported coal 638 combustion (23%) and vehicle emissions (21%) from southeast, as well as biomass burning emissions (19%) from the northeast, were also significant. Meteorological 639 640 conditions, including wind patterns and relative humidity, also influenced the regional transport of BCc particles in the YRD. 641 642 It should be noted that the observed average PM2.5 concentration during the lockdown 643 in Yangzhou was 21.2 µg m<sup>-3</sup>, which still significantly exceeds the WHO's air quality 644 645 guideline of 5 µg m<sup>-3</sup>. Our research highlights that reduction of local primary emissions from traffic and manufacturing sectors alone has limited effect in air quality 646 remediation. Complex chemistry, regional transport and meteorological factors need to 647 be considered cooperatively. Therefore, we suggest a more comprehensive regulation 648 of precursor gases from multiple sectors, a wide-ranging joint regulation approach as 649 well as proper consideration of the chemistry, so as to develop an effective strategy for 650 651 air quality improvement. 652 653 Data availability. Data described in this manuscript can be accessed at repository under: https://doi.org/10.6084/m9.figshare.24427795 (Dai, 2023) 654 655 656 Author contributions. XG and YD designed the research. YD, HW and SC conducted 657 the field measurements. YD, HW, JW and SC analyzed the data. XG, JW, HL, YW, YZ, 658 and EA reviewed the paper and provided useful suggestions. YD and XG wrote the first 659 draft of paper. All people involve in discussion of the results. 660 Supplement. The supplement related to this article is available online at: XXX. 661 662 663 Competing interests. The contact author has declared that neither they nor their co-664 authors have any competing interests. 665 666 Financial support. This research has been supported by the National Natural Science 667 Foundation of China (grant nos. 42377100 and 42021004).





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