

1 **Regional severe particle pollution and its association with**
2 **synoptic weather patterns in the Yangtze River Delta region,**
3 **China**

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13
14 **Abstract:** Regional air pollution is significantly associated with dominant weather systems. In this
15 study, the relationship between the particle pollution over the Yangtze River Delta (YRD) region
16 and weather patterns is investigated. First, the pollution characteristics of particles in the YRD are
17 studied using in situ monitoring data (PM_{2.5} and PM₁₀) in 16 cities and Terra/MODIS AOD
18 (aerosol optical depth) products collected from December 2013 to November 2014. The results
19 show that the regional mean value of AOD is high in the YRD, with an annual mean value of
20 0.71±0.57. The annual mean particle concentrations in the cities of Jiangsu Province all exceed the
21 national air quality standard. The pollution level is higher in inland areas, and the highest
22 concentrations of PM_{2.5} and PM₁₀ are 79 and 130 µg·m⁻³, respectively, in Nanjing. The PM_{2.5}/PM₁₀
23 ratios are typically high, thus indicating that PM_{2.5} is the overwhelmingly dominant particle
24 pollutant in the YRD. The wintertime peak of particle concentrations is tightly linked to the
25 increased emissions during the heating season, as well as adverse meteorological conditions.
26 Second, based on NCEP reanalysis data, synoptic weather classification is conducted, and five
27 typical synoptic patterns are objectively identified. Finally, the synthetic analysis of
28 meteorological fields and backward trajectories are applied to further clarify how these patterns
29 impact particle concentrations. It is demonstrated that air pollution is more or less influenced by

30 high-pressure systems. The relative position of the YRD to the anti-cyclonic circulation exerts
31 significant effects on the air quality of the YRD. The YRD is largely influenced by polluted air
32 masses from the northern and the southern inland areas when it is located at the rear of the East
33 Asian major trough. The significant downward motion of air masses results in stable weather
34 conditions, thereby hindering the diffusion of air pollutants. Thus, this pattern is quite favorable
35 for the accumulation of pollutants in the YRD, resulting in higher regional mean PM_{10}
36 ($116.5 \pm 66.9 \mu\text{g}\cdot\text{m}^{-3}$), $PM_{2.5}$ ($75.9 \pm 49.9 \mu\text{g}\cdot\text{m}^{-3}$) and AOD (0.74) values. Moreover, this pattern is
37 also responsible for the occurrence of most large-scale regional $PM_{2.5}$ (70.4%) and PM_{10} (78.3%)
38 pollution episodes. High wind speed and clean marine air masses may also play important roles in
39 the mitigation of pollution in the YRD. Especially when the clean marine air masses account for a
40 large proportion of all trajectories (i.e., when the YRD is affected by the cyclonic system or
41 oceanic circulation), the air in the YRD has a smaller chance of being polluted. The observed
42 correlation between weather patterns and particle pollution can provide valuable insight into
43 making decisions about pollution control and mitigation strategies.

44 **Keywords:** $PM_{2.5}$; PM_{10} ; air pollution meteorology; synoptic weather pattern; the Yangtze River
45 Delta region

46

47 **1. Introduction**

48 The common occurrence of regional particle pollution has acquired worldwide attention in
49 the scientific community (Malm et al., 1994; Putaud et al., 2004; Chan and Yao, 2008) due to its
50 adverse impacts on visibility (Singh and Dey, 2012; Green et al., 2012) and public health (Kappos
51 et al., 2004; Brook et al., 2010). Generally, the causes of this kind of pollution involve diverse
52 aspects. Two major contributors to this pollution include the emission of pollutants and weather
53 conditions (Oanh and Leelasakultum, 2011; Young et al., 2016). Particle pollution in urban
54 agglomerations is primarily attributed to very large amounts of the anthropogenic emissions of
55 primary particles and their precursors (e.g., SO_2 , NO_x , VOCs). However, these emissions are
56 normally quasi-stable within a certain period of time (Kurokawa et al., 2013). Thus, the pollution
57 level in a certain region generally depends on the regional weather conditions (namely, weather
58 patterns), which are strongly correlated with synoptic-scale atmospheric circulation (Buchanan et

59 al., 2002; Chuang et al., 2008; Flocas et al., 2009; Zhang et al., 2012; Zhao et al., 2013; Russo et
60 al., 2014; Grundstrom et al., 2015; Zheng et al., 2015a; 2015b; Li et al., 2016).

61 To date, researchers have gained an improved knowledge of the relationship between weather
62 patterns and particle pollution. For example, Buchanan et al. (2002) observed significantly
63 elevated concentrations of Black Smoke and PM₁₀ under the anti-cyclonic, southerly and
64 southeasterly weather types in the city of Edinburgh in the UK between 1981 and 1996. Russo et
65 al. (2014) presented an objective classification scheme for the atmospheric circulation affecting
66 Portugal between 2002 and 2010 and revealed that higher concentrations of PM₁₀, O₃ and NO₂ are
67 predominantly associated with synoptic circulation that is characterized by an eastern component
68 and the advection of dry air masses. Previous studies have confirmed that different levels of air
69 pollution are closely related with weather patterns, and they ascribed its great spatial variability to
70 the fact that the dominant weather pattern differs between different regions (Flocas et al., 2009;
71 Grundstrom et al., 2015).

72 In recent decades, the air pollution caused by PM₁₀ and PM_{2.5} has become an extremely
73 prominent air quality problem in the urban areas of China (Deng et al., 2011; Huang et al., 2012;
74 Ji et al., 2012; Cheng et al., 2013; Kang et al., 2013; Huang et al., 2014; Zhang et al., 2014; Xie et al.,
75 2016a; 2016c; Zhu et al., 2017). Many studies have tried to reveal the meteorological
76 contributions to these severe particle pollution episodes. Chuang et al. (2008) identified seven
77 weather patterns for aerosol events occurring from March 2002 to February 2005 in the Taipei
78 Basin and suggested that weather systems and their associated terrain blocking played important
79 roles in the accumulation of PM_{2.5} during the days of events. Niu et al. (2010) revealed the
80 potential impacts of the weakening of the East Asian monsoon circulation and increased aerosol
81 loading on the increase in wintertime fog in China. Zhao et al. (2013) analyzed a regional haze
82 episode in the North China Plain from 16 to 19 January 2010 and noted that strong temperature
83 inversion, weak surface wind speed and descending air motions in the boundary layer were
84 responsible for the accumulation of pollutants in a shallow layer that produced high pollutant
85 concentrations within the source region. Zheng et al. (2015a) found that favorable atmospheric
86 circulation conditions are responsible for the severe winter haze over northeastern China. Li et al.
87 (2016) noted that the fog-haze days over central and eastern China exhibited the clear features of

88 inter-annual variations and that the strong (weak) East Asian winter monsoon may result in less
89 (more) fog-haze days throughout this region.

90 The Yangtze River Delta (YRD) region, which is located in the southeastern coastal area of
91 East China, is one of the most developed urban economic regions in the world; it generally
92 includes Shanghai, Jiangsu Province and Zhejiang Province, and it occupies over 20% of China's
93 total gross domestic product (GDP) (Shu et al., 2016; Xie et al., 2016a; 2017). In recent years,
94 similar to other megacity clusters in China, such as the Beijing-Tianjin-Hebei (BTH) region (He et
95 al., 2001; Chan and Yao, 2008; Ji et al., 2012; Zhang et al., 2012; 2014; Zhao et al., 2013; Zheng
96 et al., 2015a) and the Pearl River Delta (PRD) region (Ho et al., 2003; Chan and Yao, 2008; Xie et
97 al., 2016c; Zhu et al., 2017), the YRD has suffered from severe air pollution problems caused by
98 an increasing population, urban expansion, and industrialization (Chan and Yao, 2008; Fu et al.,
99 2008; 2010; 2014; Deng et al., 2011; Li et al., 2011; Huang et al., 2012; Kang et al., 2013; Wang et
100 al., 2013; 2014; 2015; Xie et al., 2014; 2016a, 2016b, 2017; Feng et al., 2015; Zheng et al., 2015b;
101 Shu et al., 2016; Xu et al., 2016; Ming et al., 2017). In particular, severe particle pollution
102 episodes are widely recognized as one of the major air pollution issues in the YRD (Fu et al., 2008;
103 2010; Deng et al., 2011; Huang et al., 2012; Kang et al., 2013; Kong et al., 2013; Wang et al.,
104 2013; 2014; 2015; Fu et al., 2014; Feng et al., 2015; Zheng et al., 2015b; Xu et al., 2016; Ming et
105 al., 2017). Thus, many studies have been conducted to determine the contamination status (Fu et
106 al., 2010; Kang et al., 2013; Wang et al., 2013; 2015; Feng et al., 2015; Ming et al., 2017),
107 possible source (Fu et al., 2010; 2014; Kong et al., 2013; Wang et al., 2013; 2014; Xu et al., 2016),
108 and causes or features (Fu et al., 2008; 2010; Huang et al., 2012; Wang et al., 2015; Zheng et al.,
109 2015a) of these episodes. However, studies that have attempted to determine how particle
110 pollution in the YRD is associated with synoptic weather patterns are still quite limited. Zheng et
111 al. (2015b) summarized the synoptic-scale atmospheric circulations influencing the distribution of
112 particles over eastern China during autumn from 2001 to 2010. They found that there are six
113 polluted weather types and three clean ones and revealed that heavy pollution events most
114 commonly occur when the study areas are located at the rear of the anticyclone. However, their
115 study considered the influence of pollution in a region that is larger than YRD, only focused on
116 pollution in October, and was mainly based on satellite aerosol optical depth (AOD) data.
117 Ground-based monitoring particle concentration data can better represent the status of particle

118 pollution in the urban atmosphere of the YRD. Thus, to better understand the relationship between
119 pollution in the planetary boundary layer and the synoptic weather patterns over the YRD, further
120 studies should be conducted based on surface monitoring data collected over a time period of at
121 least one year in the YRD.

122 This work attempts to enhance our understanding of particle pollution in the YRD and
123 provide scientific knowledge about the association of regional severe particle pollution and
124 synoptic weather patterns. First, we analyze the spatial and temporal distribution of PM₁₀, PM_{2.5}
125 and AOD in the YRD from December 2013 to November 2014 to illustrate the characteristics of
126 particle pollution over this region. Second, synoptic weather classification is conducted to reveal
127 the weather patterns related to heavy pollution. Finally, the synthetic analyses of meteorological
128 fields and backward trajectories are used to further clarify the impact mechanism. In this paper,
129 Section 2 describes the observed data, the synoptic weather classification method and the
130 trajectory model. Section 3 presents our main findings, including a detailed analysis of the
131 characteristics of particle pollution in the YRD, the synoptic weather patterns affecting this
132 pollution, and the mechanism by which weather systems impact pollution. Finally, a brief
133 summary is presented in Section 4.

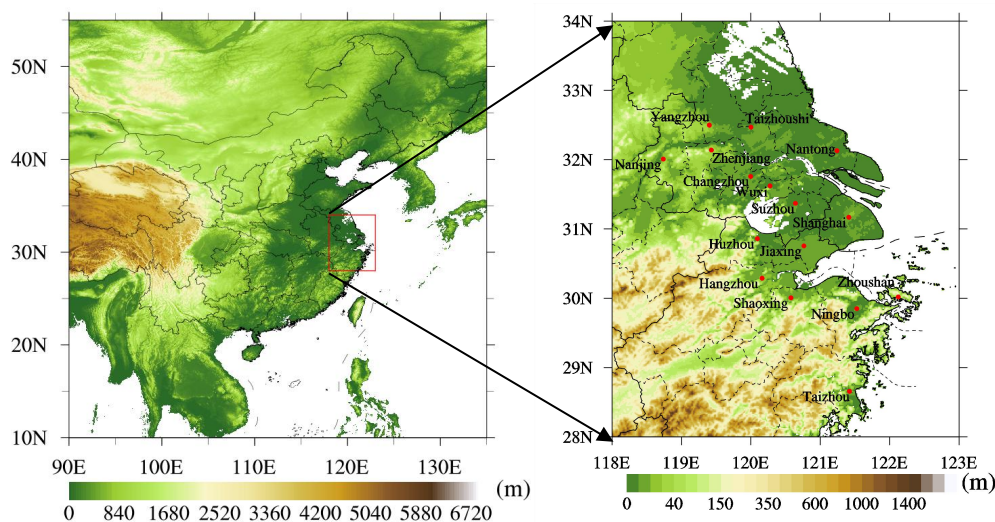
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135 **2. Data and methods**

136 **2.1 Observed data**

137 The observed air quality data used in this study are obtained from the National
138 Environmental Monitoring Center (NEMC) of China. The in situ monitoring data of the hourly
139 concentrations of PM_{2.5}, PM₁₀, CO, NO₂, SO₂ and O₃ are acquired from the national air quality
140 real-time publishing platform (<http://106.37.208.233:20035>). Sixteen cities are selected as
141 representative research sites to better reflect the status of particle pollution over the YRD region.
142 These cities include Shanghai, Changzhou, Nanjing, Nantong, Suzhou, Taizhoushi, Wuxi,
143 Yangzhou, Zhenjiang, Hangzhou, Huzhou, Jiaxing, Ningbo, Shaoxing, Taizhou, and Zhoushan
144 (here, Taizhou in Jiangsu Province is referred to as Taizhoushi to distinguish it from the city of
145 Taizhou in Zhejiang Province). Fig. 1 shows the locations of the 16 cities in the YRD. In order to
146 better characterize the pollution levels of each city, the hourly pollutant concentration of each city
147 is calculated as the average value of the pollutant concentrations measured in several of the

148 national monitoring sites in that city. The sampling methods and the quality assurance and quality
 149 control (QA/QC) procedures used at each site are in accordance with the Chinese national
 150 standard HJ/T193-2005 (State Environmental Protection Administration of China, 2006; Xie et al.,
 151 2016b). Furthermore, manual inspection is conducted during data processing; this inspection
 152 includes the removal of missing and abnormal values (e.g., $PM_{2.5}$ values that are higher than PM_{10}
 153 values). The study period lasts from December 2013 to November 2014. In the following analysis,
 154 winter refers to the period from December 2013 to February 2014. Accordingly, spring, summer
 155 and fall represent the periods from March to May, June to August, and September to November
 156 2014, respectively.
 157



158
 159
 160 **Figure 1. The location of the YRD in China (a) and 16 typical cities in the YRD (b), with terrain elevation**
 161 **data. The terrain elevation data are obtained from the website**
 162 **(https://www.ngdc.noaa.gov/mgg/global/relief/ETOPO1/data/bedrock/cell_registered/).**
 163

164 The use of Moderate Resolution Imaging Spectroradiometer (MODIS) aerosol products can
 165 help us comprehensively analyze the spatial and temporal variations in aerosol loading over China.
 166 In this study, we use the aerosol optical depth (AOD) data obtained at a wavelength of 550 nm in
 167 the Terra/MODIS daily global Level 3 products (MOD08_D3). These data can be obtained from
 168 the MODIS collection 6 (C6) dataset (<https://ladsweb.nascom.nasa.gov/search/index.html>).
 169 MODIS aerosol products are derived using two entirely independent retrieval algorithms: one is
 170 used for deriving aerosols over land (Chu et al, 2002; 2003) and another is used for deriving
 171 aerosols over the ocean (Remer et al, 2002; 2005; Chu et al., 2005). Here, we use the C6 Deep

172 Blue (DB) products to derive aerosols over land, with a spatial resolution of $1^\circ \times 1^\circ$, during the
173 period from December 2013 to November 2014. For detailed descriptions of the retrieval
174 algorithms and their accuracy and validation, refer to the work of Hsu et al. (2013).

175 To illustrate actual weather situations, the hourly monitored meteorological parameter
176 records in each of the 16 typical cities are also applied. These data include 2 m temperature (T), 2
177 m relative humidity (RH), 10 m wind speed (WS), 10 m wind direction (WD) and surface air
178 pressure (P). These data are collected from the National Meteorological Center
179 (<http://www.nmc.cn>).

180

181 2.2 Synoptic weather classification

182 Synoptic weather classification refers to the analysis of historical weather charts and the
183 characterization of weather systems. It is more effective for producing disastrous weather forecasts
184 due to its ability to reveal atmospheric circulation situations. With the gradual popularization of
185 computer analysis and the increased sharing of data, synoptic weather classification has great
186 practical value in a wide variety of research fields. For example, it has widespread applications in
187 the field of analyzing weather patterns related to air pollution (Mcgregor and Bamzeli, 1995;
188 Zhang et al., 2012; Santurtún et al., 2015).

189 Methods of synoptic weather classification can generally be divided into objective and
190 subjective methods (El-Kadi and Simithson, 1992). In this study, we apply the sums-of-squares
191 technique, which is an objective classification method that was established in 1973 by Kirchhofer
192 (Kirchhofer, 1973). The sums-of-squares technique can effectively categorize more than 90% of
193 analyzed weather maps, which represents an improvement over other correlation techniques
194 (Yarnal, 1984). The application of this technique involves three steps. First, the daily pressure data
195 at each grid point are normalized as follows:

$$196 \quad Z_i = \frac{(X_i - \bar{X})}{s} \quad (1)$$

197 where Z_i is the normalized value of grid point i , X_i is the value at grid point i , \bar{X} is the mean
198 value of the study domain, and s is the standard deviation. Data normalization removes the effects
199 of the magnitude of pressure and improves the seasonal comparability of different weather types.
200 Second, each normalized grid point is compared to all other grid points based on the Kirchhofer

201 score (S) of each grid point:

$$202 \quad S = \sum_{i=1}^N (Z_{ai} - Z_{bi}) \quad (2)$$

203 where Z_{ai} is the normalized value of grid point i on day a , Z_{bi} is the normalized value of grid point
204 i on day b , and N is the number of grid points. The Kirchofer score (S) is calculated for each row
205 (denoted as S_R), each column (S_C) and the entire study domain (S_T) to ensure the pattern similarity
206 between any pair of patterns for all grid points. Finally, all days are separated into one of the
207 identified synoptic weather patterns based on these three values and their empirically derived
208 thresholds. Thus, the values of S_R , S_C and S_T must be lower than their respective threshold values
209 for these patterns to be accepted as similar (Barry et al., 1981). For each daily grid, the lowest
210 significant Kirchofer score (S) is recorded with the associated key day, thus denoting the
211 synoptic type of that day. All remaining days are considered to be ‘unclassified’.

212 The meteorological field dataset used in the sums-of-squares technique contains NCEP–DOE
213 AMIP-II Reanalysis 2 data (Kanamitsu et al., 2002), which are collected at 00:00, 06:00, 12:00,
214 and 18:00 UTC (universal time coordinated)
215 (<https://www.esrl.noaa.gov/psd/data/gridded/data.ncep.reanalysis2.pressure.html>). These data have
216 144×73 horizontal grids with a grid spacing of 2.5° . From the ground level to 10 hPa, there are 17
217 pressure levels in the vertical direction. The classification of synoptic weather maps is conducted
218 using the gridded data at a geopotential height of 850 hPa during the same time period when the
219 air quality data are recorded. The domain of interest is centered over the YRD region, covering an
220 area of $25\text{--}40^\circ$ N in latitude and $110\text{--}128^\circ$ E in longitude.

221

222 **2.3 HYSPLIT model**

223 Backward trajectories can be adopted to help understand transport paths and identify the
224 source regions of air masses. The Hybrid Single-Particle Lagrangian Integrated Trajectory
225 (HYSPLIT) Model (Version 4) was developed by the National Oceanic and Atmospheric
226 Administration (NOAA) Air Resources Laboratory (ARL). It is one of the most extensively used
227 atmospheric transport and dispersion models for the study of air parcel trajectories (Draxler and
228 Rolph, 2013; Rolph, 2013; Stein et al., 2016), and it has been widely applied in simulations of the
229 complex transport, diffusion, chemical transformation and deposition processes of atmospheric

230 pollutants (Mcgowan and Clark, 2008; Wang et al., 2011; Huang et al., 2015; Xie et al., 2016b).

231 In this study, HYSPLIT is used to compute the backward trajectories of air parcels, reveal the
232 possible source regions of air masses, and establish source-receptor relationships for each synoptic
233 weather pattern. For each synoptic weather pattern, the terminus of each trajectory is considered to
234 be located at the observation site in Nanjing (32°N, 118.8°E). The 72-h backward trajectories are
235 then calculated and clustered. The ending point is defined as 1500 m above sea level. The NCEP
236 reanalysis data (<http://ready.arl.noaa.gov/archives.php>) are used to drive the backward trajectory
237 calculation. The NCEP data contain 6-hourly basic meteorological fields on pressure surfaces with
238 a spatial resolution of 2.5°. In this study, these data are also converted to hemispheric 144 by 73
239 polar stereographic grids; these data thus have the same grid configuration as the dataset applied
240 in the synoptic weather classification.

241

242 **3. Results and discussion**

243 **3.1 Characteristics of particle pollution in the YRD**

244 **3.1.1 Spatial distributions of particle pollution**

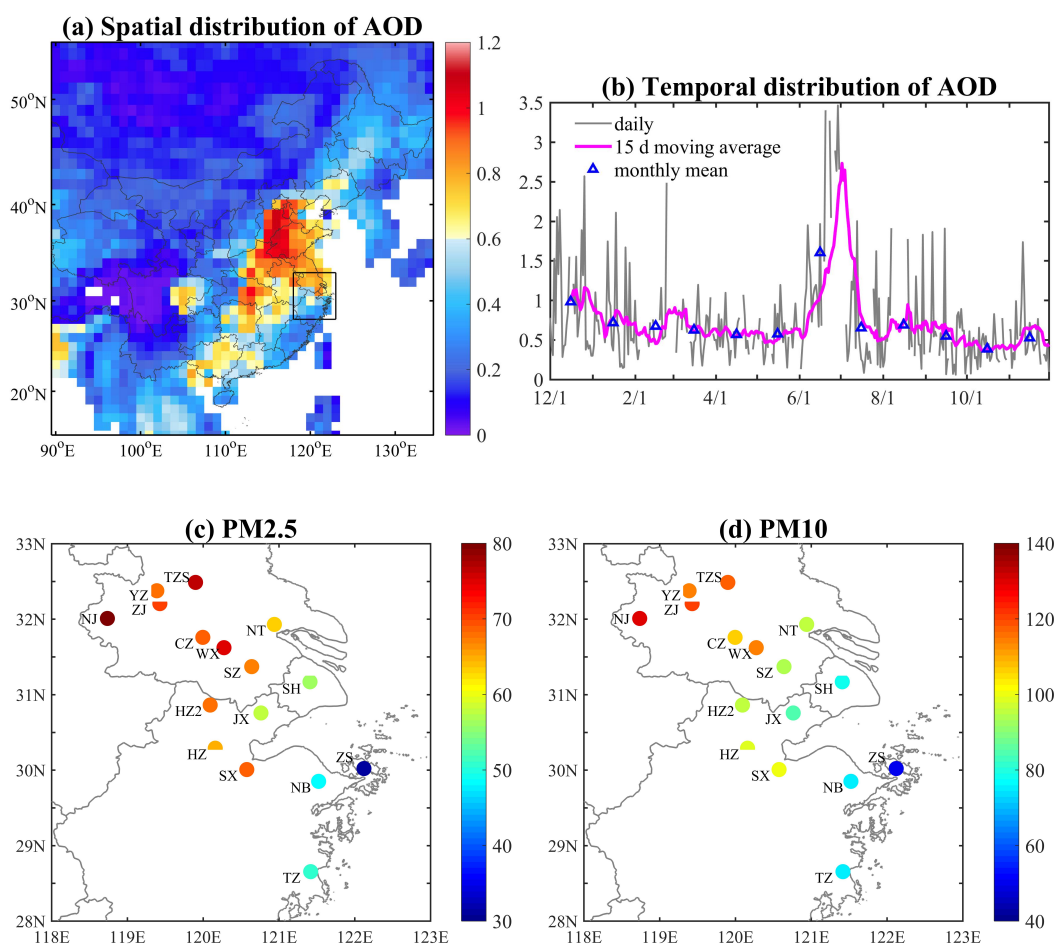
245 Fig. 2a displays the annual mean values of AOD observed at a wavelength of 550 nm
246 throughout most of China. The highest values (i.e., larger than 0.6) generally occur in the BTH,
247 the YRD, the Sichuan Basin (SCB), and some of the central and southern provinces in China (i.e.,
248 Hubei, Hunan and Guangxi provinces). AOD is mainly governed by fine particles in industrialized
249 urban conditions (Kim et al., 2006); thus, the abovementioned areas should suffer from high
250 columnar aerosol loading. In the YRD, with the development of modern industrialization and
251 urbanization, contrasts in the atmospheric pollution levels among different cities gradually
252 decrease, and severe air pollution episodes tend to exhibit significant regional pollution
253 characteristics.

254 Fig. 2b shows the temporal variations in the regional average AOD values in the YRD
255 (covering 16 cities within the area of 25-40°N and 110-128°N). The annual mean value is
256 0.71 ± 0.57 . The maximum seasonal value is 0.98 ± 0.83 in summer, followed by 0.81 ± 0.57 in winter,
257 0.59 ± 0.24 in spring, and 0.48 ± 0.35 in autumn. Although the peak particle concentrations are
258 observed in winter (as shown in Fig. 3 and 5), the above results demonstrate that the maximum
259 regional mean AOD values occur in summer, as they reach their highest value of 1.60 in June.

260 This result is similar to that found by Kim et al. (2006), who reported that the value of AOD is not
261 only associated with the pollution levels of fine particles but is also strongly affected by other
262 factors (e.g., solar radiation, water vapor). The fact that the maximum AOD values occur in hot
263 seasons should be ascribed to the combined effects of the increase in fine aerosol production (i.e.,
264 due to secondary aerosol formation by gas-to-particle conversion, the hygroscopic growth of
265 hydrophilic aerosols or biomass burning emissions) and humid weather (Kim et al., 2006).
266 Consequently, the aerosol optical depth data obtained from satellite observations can reveal the
267 spatial distribution of aerosols to some extent, but they cannot exactly reflect pollution levels or
268 replace concentration data.

269 Figs. 2c and 2d show the spatial distributions of the annual mean particle concentrations in 16
270 typical cities over the YRD from December 2013 to November 2014. Generally, the spatial
271 distributions of PM_{2.5} (Fig. 2c) and PM₁₀ (Fig. 2d) exhibit overall similar patterns. The annual
272 mean PM_{2.5} and PM₁₀ values decrease progressively in the northwest-southeast direction, which
273 means that particle concentrations are comparatively high in the northwest inland areas and low in
274 the southeast coastal areas. The pollution levels in most cities exhibit a positive correlation with
275 their proximity to the sea. The farther a city is from the sea, the higher its particle concentrations
276 are. The maximum particle concentrations occur in Nanjing, with values of 79 $\mu\text{g}\cdot\text{m}^{-3}$ for PM_{2.5}
277 and 130 $\mu\text{g}\cdot\text{m}^{-3}$ for PM₁₀. Previous studies of major climatic features in the YRD have
278 demonstrated that the southeast coastal area is dramatically affected by the land-sea breeze and
279 marine air masses. The clean marine air masses are advantageous to the dilution and diffusion of
280 atmospheric pollutants, thus producing lighter air pollution. However, in the inland region,
281 clustered cities and industrial districts tend to emit more pollutants, thereby resulting in the
282 accumulation of more air pollutants around these cities.

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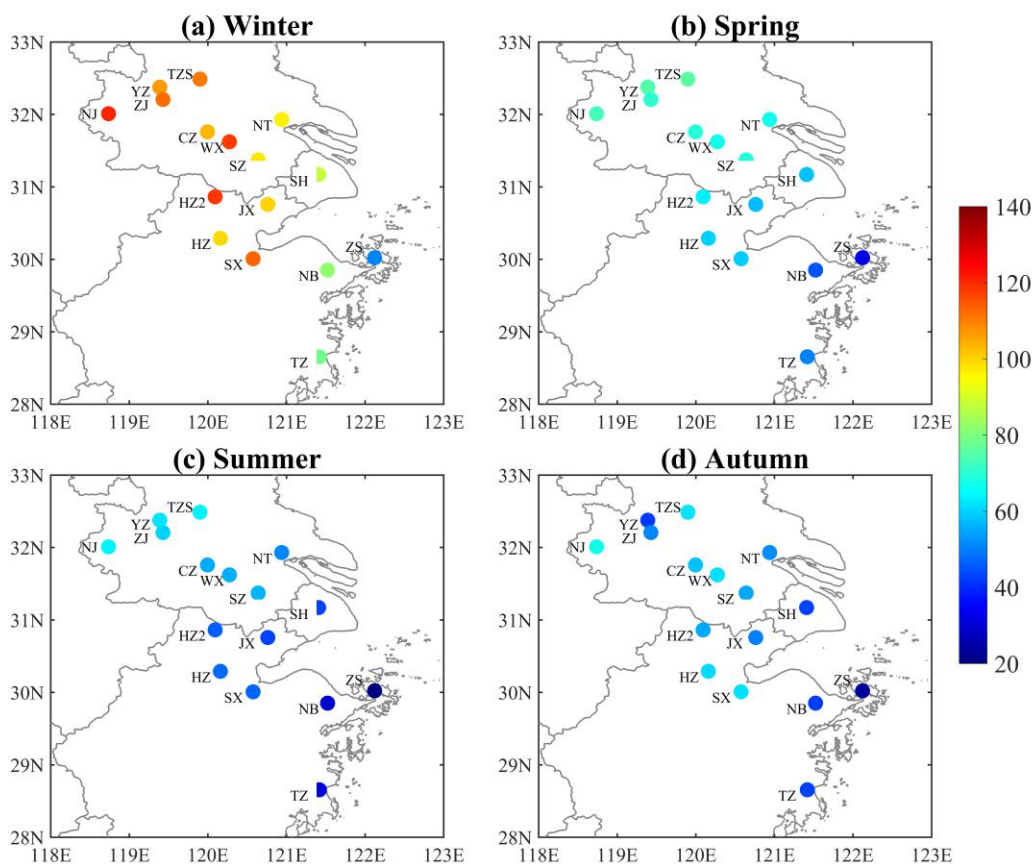
286

287 **Figure 2.** The spatial distribution of annual mean AOD values (at a wavelength of 550 nm) over the
 288 YRD (a); the temporal variations in regional average AOD values over 28-33°N and 118-123°E (b); the
 289 spatial distribution of annual mean PM_{2.5} concentrations (c); and the spatial distribution of annual mean
 290 PM₁₀ concentrations (d). In (b), the gray line represents the daily value, the blue markers represent the
 291 monthly mean values, and the magenta line represents the 15-day moving average value. In (c) and (d), the
 292 acronyms of each city are marked, including Shanghai-SH, Changzhou-CZ, Nanjing-NJ, Nantong-NT,
 293 Suzhou-SZ, Taizhoushi-TZS, Wuxi-WX, Yangzhou-YZ, Zhenjiang-ZJ, Hangzhou-HZ, Huzhou-HZ2,
 294 Jiaying-JX, Ningbo-NB, Shaoxing-SX, Taizhou-TZ, and Zhoushan-ZS.

295

296 Fig. 3 illustrates the spatial distribution of the seasonal mean PM_{2.5} in 16 cities over the YRD.
 297 The pattern observed during each season is similar to the annual mean pattern (Fig. 2c). The PM_{2.5}
 298 pollution levels are much higher in inland cities, and they decrease in the northwest-southeast
 299 direction. PM_{2.5} concentrations exhibit seasonal variations; they are highest in winter, reaching a
 300 maximum value of 120 µg·m⁻³, and they decrease throughout spring, yielding their lowest values
 301 during summer and autumn. The difference between the PM_{2.5} concentration in summer and that
 302 in autumn is relatively small; this difference ranges from a maximum value of lower than 60

303 $\mu\text{g}\cdot\text{m}^{-3}$ in Nanjing to a minimum value of close to $20 \mu\text{g}\cdot\text{m}^{-3}$ in Zhoushan.



304
 305 **Figure 3. The spatial distribution of seasonal mean $\text{PM}_{2.5}$ over the YRD in (a) winter, (b) spring, (c) summer,**
 306 **and (d) autumn (unit: $\mu\text{g}\cdot\text{m}^{-3}$). The acronyms for each city are the same as those in Figure 2.**

307
 308 Table 1 quantitatively lists the annual mean concentrations of $\text{PM}_{2.5}$ and PM_{10} in 16 cities
 309 over the YRD. It also demonstrates that the particle pollution levels are relatively higher in inland
 310 cities. The concentrations of $\text{PM}_{2.5}$ and PM_{10} in 8 cities in Jiangsu Province are all higher than 60
 311 $\mu\text{g}\cdot\text{m}^{-3}$ ($\text{PM}_{2.5}$) and 80 $\mu\text{g}\cdot\text{m}^{-3}$ (PM_{10}), respectively. However, these concentrations are
 312 comparatively lower in the cities located in the coastal area (e.g., Ningbo, Taizhou and Zhoushan).
 313 Only the air quality of Zhoushan meets the national standard, which may be attributed to the fact
 314 that it is located on an island, where its air is most likely influenced by clean marine air masses.

315 To reveal the important role of $\text{PM}_{2.5}$ in particle pollution, the ratios of $\text{PM}_{2.5}$ concentration to
 316 PM_{10} concentration ($\text{PM}_{2.5}/\text{PM}_{10}$) are calculated over the YRD. As listed in Table 1, the maximum
 317 annual mean value of the $\text{PM}_{2.5}/\text{PM}_{10}$ ratio is 0.72 in Shanghai, followed by Huzhou and Suzhou
 318 (0.71), thus implying that the $\text{PM}_{2.5}$ fraction is overwhelmingly dominant relative to the PM_{10}
 319 mass in these cities. The $\text{PM}_{2.5}/\text{PM}_{10}$ ratios in other cities range from 0.60 to 0.69, with a

320 minimum value of 0.58 in Zhenjiang. These values are comparable to those in other cities, such as
321 Beijing (He et al., 2001), Shanghai (Wang et al., 2013), Taipei (Chen et al., 1999), and Hong Kong
322 (Ho et al., 2003), thus suggesting that the formation of PM_{2.5} from gases is the most important
323 source of particles in the cities of China. Table 1 also indicates that the PM_{2.5}/PM₁₀ ratios in all
324 cities exhibit distinct seasonal variation. It is remarkable that the values of PM_{2.5}/PM₁₀ are much
325 higher in winter than they are in other seasons, reaching a maximum value of 0.85 in Shanghai,
326 followed by a value of 0.82 in Suzhou. The highest concentrations of PM_{2.5} usually occur in
327 winter (Fig. 3a), and high values of the PM_{2.5}/PM₁₀ ratio also occur during the same season (Table
328 1), thus indicating that PM_{2.5} poses a greater threat to human health in cold seasons, which may be
329 related to heating activities. In summer, the values of PM_{2.5}/PM₁₀ in the 16 cities are medium, with
330 a mean value of 0.67. The lowest ratios usually occur in spring and autumn, when the mean ratios
331 of all cities are 0.61 (spring) and 0.63 (autumn). The minimum value occurs in the autumn in
332 Yangzhou, with a value of 0.51, followed by a value of 0.52 in the spring in Nanjing and the
333 autumn in Zhenjiang. The above discussion of the spatial and temporal variations in PM_{2.5}/PM₁₀
334 ratios also implies that particles originate from various kinds of sources and are variedly emitted.

335

336 **Table 1. Annual mean concentrations of PM_{2.5} and PM₁₀, and the annual and seasonal mean values of PM_{2.5}/
337 PM₁₀ ratio, in 16 cities over the YRD.**

Cities	PM _{2.5} ($\mu\text{g}\cdot\text{m}^{-3}$)	PM ₁₀ ($\mu\text{g}\cdot\text{m}^{-3}$)	PM _{2.5} /PM ₁₀					
			Annual	Winter	Spring	Summer	Autumn	
Shanghai	56	78	0.72	0.85	0.68	0.72	0.66	
Nanjing	79	130	0.61	0.64	0.52	0.70	0.60	
Changzhou	69	106	0.65	0.73	0.60	0.67	0.62	
Nantong	63	95	0.66	0.72	0.62	0.71	0.64	
Jiangsu Province	Suzhou	67	94	0.71	0.82	0.68	0.71	0.67
Taizhoushi	76	117	0.65	0.66	0.58	0.72	0.66	
Wuxi	75	114	0.66	0.73	0.59	0.67	0.62	
Yangzhou	68	114	0.60	0.69	0.58	0.59	0.51	
Zhenjiang	70	121	0.58	0.71	0.54	0.58	0.52	
Hangzhou	65	99	0.66	0.74	0.59	0.63	0.66	
Zhejiang Province	Huzhou	68	96	0.71	0.78	0.66	0.68	0.69
Jiaxing	58	84	0.69	0.75	0.65	0.68	0.69	
Ningbo	48	75	0.64	0.69	0.62	0.63	0.62	
Shaoxing	68	100	0.68	0.72	0.62	0.71	0.68	

Taizhou	50	75	0.67	0.69	0.66	0.66	0.65
Zhoushan	31	50	0.63	0.66	0.62	0.66	0.55

338

339 **3.1.2 Temporal variations in particle pollution**

340 Fig. 4 shows the annual mean diurnal variations in PM_{2.5} (Fig. 4a) and PM₁₀ (Fig. 4b) in 16
341 cities over the YRD. Obviously, the diurnal cycles of particle concentrations in most cities follow
342 a similar pattern. The PM_{2.5} concentrations maintain comparably high values from 0:00 to 8:00.
343 Then, coinciding with more vehicle emissions during rush hours, these concentrations increase
344 rapidly from 8:00 to 12:00. After reaching their peak, the PM_{2.5} concentrations decrease and
345 remain at low values until sunset. During nighttime, the pollutants accumulate until midnight,
346 which can be attributed to the more stable atmospheric stratification in the boundary layer. In
347 comparison, there are two peaks in the diurnal cycles of the PM₁₀ concentrations in several cities.
348 The broad morning peak of PM₁₀ concentrations is more evident from 8:00 to 12:00, and the
349 evening peak occurs at approximately 20:00. In addition, the diurnal change in particle
350 concentrations in the southeast coastal area, such as Zhoushan, is much smaller. As discussed in
351 Section 3.1.1, this difference might be related to its special geographic location, which exhibits
352 fewer emissions of precursors and lower pollution levels.

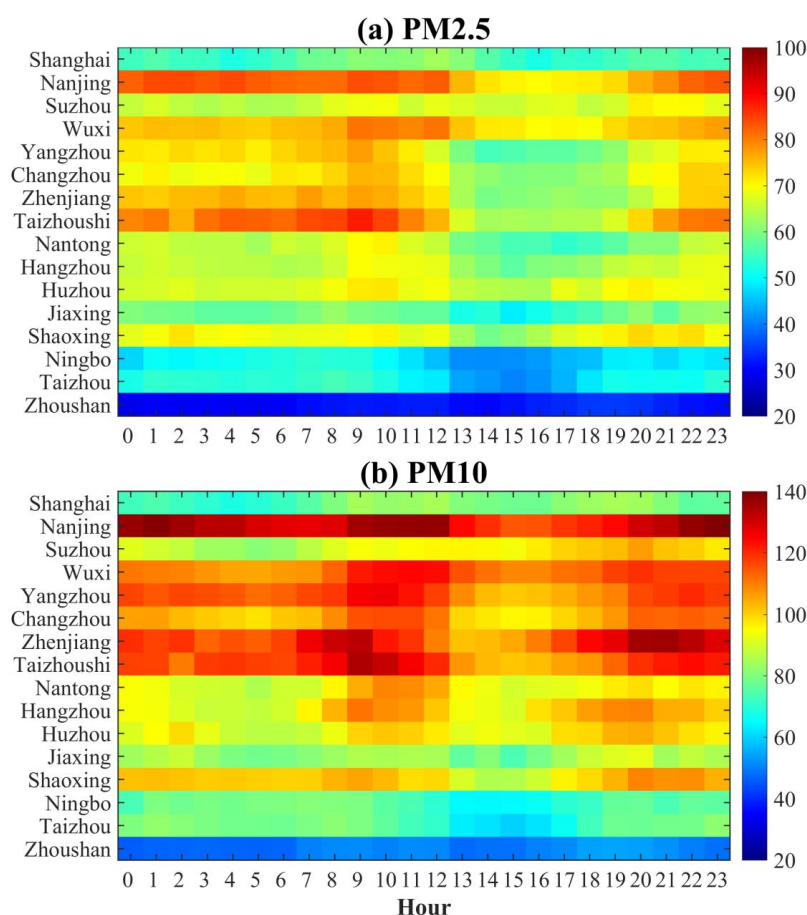


Figure 4. Diurnal variations in PM_{2.5} (a) and PM₁₀ (b) concentrations in 16 cities of the YRD (unit: µg·m⁻³).

353
354
355

356 Fig. 5 shows the monthly mean concentrations of PM_{2.5} and PM₁₀ in 16 cities of the YRD. As
357 illustrated in this figure, there are three peaks in the seasonal variations in particles. These three
358 peaks occur in December, March, and May/June. This monthly variation pattern is more obvious
359 for PM₁₀. The causes resulting in the wintertime peak of particle concentrations can be explained
360 by two factors. One is the enhanced emissions of pollutants from residential heating. The other is
361 the stable and poor meteorological conditions that limit the diffusion of atmospheric pollutants.
362 The drivers of the peak appearing in March may be associated with dust storm events in spring
363 (Zhuang et al., 2001; Fu et al., 2010; 2014). As discussed in Section 3.1.1, the values of the
364 PM_{2.5}/PM₁₀ ratio in 16 cities are lowest in spring, with a mean ratio of 0.61. High PM₁₀
365 concentrations during this period further demonstrate that dust storms can bring more coarse dust
366 particles to the YRD. The peak in May/June is probably caused by the field burning of crop
367 residue in rural areas of China, which is regarded to be an important source of biomass burning
368 (Yan et al., 2006; Yang et al., 2007; Zhu et al., 2012).

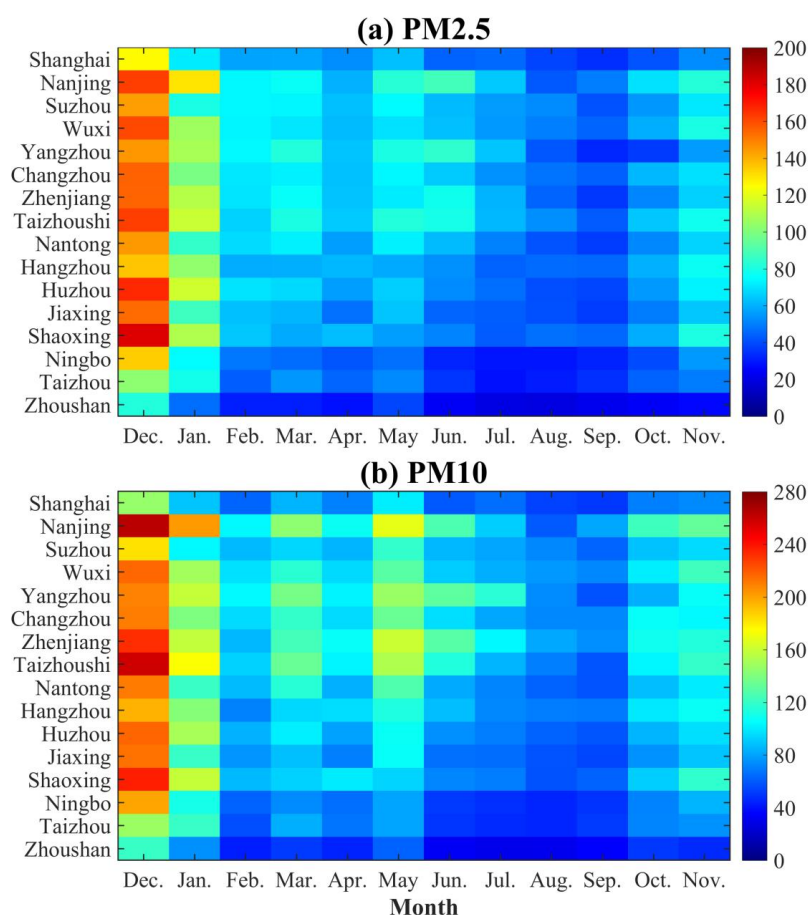


Figure 5. Monthly variations in PM_{2.5} (a) and PM₁₀ (b) concentrations in 16 cities of the YRD (unit: µg·m⁻³).

3.1.3 Regional severe particle pollution in the YRD

According to the National Ambient Air Quality Standard (NAAQS) of China, urban air quality must meet the second standard, with daily mean concentrations of PM_{2.5} and PM₁₀ that are lower than 75 µg·m⁻³ and 150 µg·m⁻³, respectively. In this study, when the daily mean PM_{2.5} (PM₁₀) concentrations exceed the national air quality standard in most (i.e., 8 or more) of the 16 cities, we define this as large-scale regional PM_{2.5} (PM₁₀) pollution. Consequently, from December 2013 to November 2014, there were 98 (46) days when large-scale regional PM_{2.5} (PM₁₀) pollution episodes were identified. That is, the YRD suffered from regional PM_{2.5} (PM₁₀) pollution during nearly 28.0% (13.1%) of the days of the year.

Table 2 shows the typical regional severe particle pollution episodes (that lasted no less than 3 days) in the YRD from December 2013 to November 2014. As illustrated in this table, dozens of continuous large-scale particle pollution episodes occurred. For example, PM_{2.5} concentrations exceeded the national standard in all 16 cities from December 1 to 5, 2013, and there were more than 14 cities facing heavy PM₁₀ pollution at the same time. From May 26 to 30, 2014, serious

386 PM_{2.5} and PM₁₀ pollution episodes were observed in more than 10 cities. It appears that
 387 high-PM_{2.5} pollution episodes are remarkably associated with high-PM₁₀ pollution episodes.
 388 Moreover, regional PM_{2.5} pollution episodes occurred much more frequently than PM₁₀ pollution
 389 episodes. This may be due to the fact that fine particles dominate the composition of particles in
 390 the YRD (as discussed in Section 3.1.2).

391

392 **Table 2. The typical regional severe particle pollution episodes (lasting for no less than 3 days) in the YRD**
 393 **from December 2013 to November 2014.**

Episodes of PM _{2.5} pollution	Episodes of PM ₁₀ pollution
1-6 Dec.	1-6 Dec.
11-15 Dec.	12-15 Dec.
24-26 Dec.	24-26 Dec.
28 Dec. - 6 Jan.	29 Dec. - 5 Jan.
15-20 Jan.	17-20 Dec.
30 Jan. - 2 Feb.	26-30 May
20-24 Feb.	
16-18 Mar.	
8-10 Apr.	
20-22 May	
26-30 May	
5-7 Jun.	
28 Jun. - 1 Jul.	
10-12 Nov.	

394

395 3.2 Synoptic weather classification

396 In this study, to examine the relationship between regional severe particle pollution in the
 397 YRD and weather situations, synoptic weather classification is carried out from December 2013 to
 398 November 2014. Using the method described in Section 2.2, we conduct the classification of the
 399 synoptic weather pattern by using the dataset of geopotential height at 850 hPa collected from the
 400 NCEP reanalysis data. As shown in Table 3, five weather patterns are finally identified. Unknown
 401 patterns are defined as ‘the unclassified pattern’. The weather situation on 95.6% of the days
 402 during the study period is classified as one of the five typical synoptic weather patterns.

403 Table 3 lists the typical date, number of days, and seasonal occurrence frequencies of each
 404 synoptic weather pattern. As demonstrated in this table, Pattern 1 is the dominant weather pattern
 405 in the YRD, which accounts for 47.6% of all of the days of the year (from December 2013 to

406 November 2014). The occurrence frequencies of Patterns 2 and 3 are 20.0% and 18.1%,
 407 respectively. Patterns 4 and 5 are identified on the fewest number of days, with occurrence
 408 frequencies of 4.1% and 5.8%, respectively.

409 Table 3 also shows the seasonal occurrence frequencies of each pattern from December 2013
 410 to November 2014. Obviously, they are distinctly different. Pattern 1 tends to occur in winter, with
 411 a frequency of 30.5%, followed by spring (25.9%), summer (21.8%) and autumn (21.8%). Pattern
 412 2 is the most popular weather pattern in summer, with an occurrence frequency of 37.0%,
 413 followed by spring (30.1%), autumn (21.9%) and winter (11.0%). For Pattern 3, the seasonal
 414 frequencies occur in the order of winter (36.4%), spring (27.3%), autumn (19.7%) and summer
 415 (16.7%). Both Pattern 4 and Pattern 5 are most likely to occur in autumn, with occurrence
 416 frequencies of 53.3% and 42.9%, respectively. The occurrence frequencies of Pattern 4 and Pattern
 417 5 during other seasons account for nearly 50%.

418

419 **Table 3. The typical date, number of days, and seasonal occurrence frequencies of each synoptic weather**
 420 **pattern.**

Type	Typical date	Number of days	Occurrence frequency (%)			
			Spring	Summer	Autumn	Winter
Pattern 1	2014-05-12	174 (47.7%)	25.9	21.8	21.8	30.5
Pattern 2	2014-05-09	73 (20.0%)	30.1	37.0	21.9	11.0
Pattern 3	2014-02-18	66 (18.1%)	27.3	16.7	19.7	36.4
Pattern 4	2014-10-07	15 (4.1%)	13.3	26.7	53.3	6.7
Pattern 5	2014-09-14	21 (5.8%)	19.0	23.8	42.9	14.3
Unclassified pattern	—	16 (4.4%)	—	—	—	—

421

422 **3.3 Effects of synoptic weather patterns on particle pollution**

423 **3.3.1 Relationship between synoptic weather pattern and particle pollution**

424 To determine the relationship between synoptic weather patterns and particle pollution, the
 425 occurrence frequencies of the five typical synoptic patterns during the regional severe particle
 426 pollution episodes are calculated. As shown in Table 4, during the days with regional PM_{2.5} (PM₁₀)
 427 pollution episodes, Pattern 1 is the dominant synoptic weather pattern, with an occurrence
 428 frequency of 70.4% (78.3%). Pattern 2 and Pattern 3 both occur on 14.3% of the days with PM_{2.5}

429 pollution episodes. During PM₁₀ pollution episodes, Pattern 2 (6.5%) appears less frequently than
 430 Pattern 3 (15.2%). The occurrence frequencies of Pattern 4 and Pattern 5 are less than 1% and can
 431 thus almost be ignored.

432 According to Table 3 and Table 4, the occurrence frequency of Pattern 1 during regional
 433 particle pollution episodes is obviously higher than its occurrence during the entire year. In
 434 contrast, the occurrences of Pattern 2 and Pattern 3 during these episodes are less frequent than
 435 those throughout the year. Moreover, Pattern 4 and Pattern 5 appear far less frequently during
 436 regional particle pollution episodes than they do throughout the year. In summary, these data
 437 suggest that the weather situation of Pattern 1 is more beneficial for the formation of large-scale
 438 regional particle pollution in the YRD.

439

440 **Table 4. The occurrence frequencies of synoptic weather patterns during regional severe PM_{2.5} and PM₁₀**
 441 **pollution episodes**

Type	PM _{2.5}		PM ₁₀	
	Number of days	Frequency (%)	Number of days	Frequency (%)
Pattern 1	69	70.4	36	78.3
Pattern 2	14	14.3	3	6.5
Pattern 3	14	14.3	7	15.2
Pattern 4	0	0%	0	0
Pattern 5	1	1.0	0	0

442

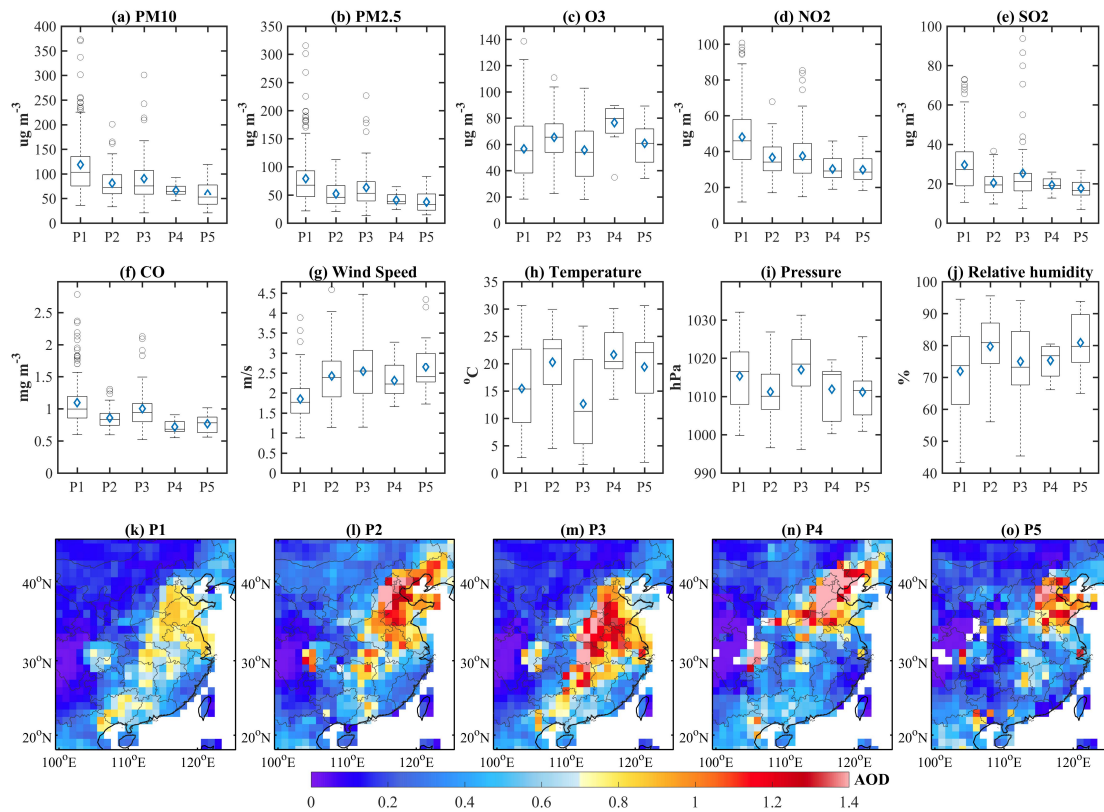
443 Fig. 6 shows the box-and-whisker plot of the mean concentrations of air pollutants (PM₁₀,
 444 PM_{2.5}, O₃, NO₂, SO₂ and CO) and the meteorological parameters (WS, T, P and RH) of 16 cities
 445 under the five synoptic weather patterns, as well as the corresponding spatial distribution of AOD
 446 over eastern China. These statistical results are also listed in Table 5.

447 As shown in Figs. 6a-6f and Table 5, the highest average concentrations of the main air
 448 pollutants (except for O₃) in the 16 cities in the YRD are associated with Pattern 1. Since aerosols
 449 can reflect and absorb solar radiation and thereby cause the photochemical production of O₃ to
 450 decrease (Kaufman et al, 2002), the O₃ concentration is lowest for Pattern 1 (Fig. 6c). As
 451 mentioned above, Pattern 1 is most likely to occur during winter (30.5%) and spring (25.9%).
 452 Therefore, the weather situation of this pattern features the weakest surface wind, lowest humidity,
 453 second-highest surface pressure, and low temperature. All of these weather characteristics are
 454 conducive to the accumulation of particles and their precursors (i.e., SO₂, NO₂ and CO). For

455 Pattern 3, the concentrations of PM₁₀, PM_{2.5}, NO₂ and SO₂ are the second-highest compared to
456 those of the other patterns. This pattern features the highest surface pressure and much stronger
457 surface wind. The temperature is the lowest, as Pattern 3 also tends to occur during winter (37.0%)
458 and spring (30.1%). Under the weather situation of Pattern 1 and Pattern 3, the YRD is both under
459 the control of high pressure and likely to suffer serious particle pollution. The strength of the
460 surface wind for different weather patterns plays a key role in the occurrence frequency of
461 regional severe particle pollution episodes. Pattern 1, which has the weakest surface wind, is
462 regarded as ‘the most polluted pattern’. The pollution levels of the main pollutants in Pattern 2 are
463 in the middle and slightly lower than those of Pattern 3. Due to its high occurrence frequency in
464 summer (37.0%) and spring (30.1), the weather condition of Pattern 2 is characterized by its
465 relatively high temperature, low pressure, and the lowest RH. In contrast, Pattern 4 and Pattern 5
466 are ‘the clean patterns’, in which the concentrations of all of their pollutants are distinctly lower
467 than those of the other three patterns. Their meteorological conditions of relatively high humidity,
468 high temperature, strong wind (especially for Pattern 5) and much lower surface pressure are also
469 favorable for the mitigation of pollutants.

470 Figs. 6k to 6o display the spatial distribution of AOD over eastern China under different
471 synoptic weather patterns. The regional mean values of AOD in the YRD (28-33°N, 118-123°N)
472 are 0.74 for Pattern 1, 0.64 for Pattern 2, 0.81 for Pattern 3, 0.47 for Pattern 4 and 0.49 for Pattern
473 5. Additionally, AOD is higher over the YRD for Pattern 3, Pattern 1 and Pattern 2. For these three
474 patterns, high AOD values usually occur in the BTH, the YRD, and the SCB, as well as the
475 provinces of Shanxi, Shandong, Hubei, Hunan, Anhui and Guangxi. The highest AOD values are
476 mainly found in northeastern China. For Pattern 4 and Pattern 5, high AOD values are mostly
477 concentrated in the BTH and Shandong Province, while relatively low AOD values are found in
478 the YRD. Since AOD is closely related to the concentrations of fine particles, it can be concluded
479 that the YRD is most heavily polluted under the weather situations of Pattern 1 and Pattern 3.

480



481

482

483 **Figure 6. (a-j) Box-and-whisker plots for the mean values of air pollutant concentrations and meteorological**
 484 **parameters of 16 typical YRD cities. The edges of each box in (a-j) are the 25th and 75th percentiles; the band**
 485 **inside the box is the median; the diamond is the average; and the whiskers extend to the most extreme data**
 486 **values. (k-p) Spatial distributions of AOD for the five synoptic weather patterns. P1, P2, P3, P4, and P5**
 487 **represent Pattern 1, Pattern 2, Pattern 3, Pattern 4, and Pattern 5, respectively.**

488

489 **Table 5. The average values of air pollutant concentrations and meteorological factors for the 16 typical**
 490 **YRD cities under different synoptic weather patterns.**

Type	PM ₁₀	PM _{2.5}	O ₃	NO ₂	SO ₂	CO	SO ₂	WS	T	P	RH
Pattern 1	116.5±66.9	75.9±49.9	57.7±27.3	46.9±19.2	29.3±17.1	1.08±0.41	29.3±17.1	1.84±0.67	15.8±7.8	1015.0±8.5	72.3±14.4
Pattern 2	81.5±38.4	52.3±27.4	65.5±23.6	36.1±13.4	20.6±9.9	0.86±0.24	20.6±9.9	2.38±0.88	20.3±6.3	1011.2±6.7	79.8±10.2
Pattern 3	86.9±49.5	59.1±37.3	58.5±25.5	35.1±15.5	23.3±15.9	0.96±0.35	23.3±15.9	2.59±0.87	13.4±8.2	1016.1±9.6	76.0±11.6
Pattern 4	66.1±18.8	40.7±15.9	76.8±19.6	29.4±9.8	19.4±6.4	0.72±0.17	19.4±6.4	2.29±0.64	21.7±4.9	1011.8±7.0	75.4±5.8
Pattern 5	58.7±31.3	37.4±22.5	61.1±20.6	29.1±11.1	17.8±8.4	0.77±0.22	17.8±8.4	2.63±0.93	19.4±8.0	1011.1±6.9	81.0±9.8

491

492 3.3.2 The impact mechanism of synoptic weather patterns on severe particle pollution

493 Figs. 7-11 present the meteorological fields and backward trajectories under the weather
 494 situations of the Pattern 1 (northwesterly inland wind), Pattern 2 (southwesterly), Pattern 3 (northerly
 495 inland wind), Pattern 4 (cyclone-related) and Pattern 5 (oceanic circulation related). The first two
 496 graphs of Figs. 7-11 illustrate the 850 hPa and 500 hPa geopotential height field and wind field,
 497 respectively. The third graphs display the sea level pressure field and 1000 hPa wind field. The

498 highlighted boxes denote the study area (i.e., the YRD). The fourth graphs demonstrate the
499 height-latitude cross-sections of vertical velocity over the latitudes of 25-40°N, which are
500 averaged from the longitudes of 110-128°E. The bold black lines show the latitude range of 16
501 cities (28.6-32.5°N) over the YRD. The positive wind speeds (10^2 Pa s^{-1}) represent vertical
502 downward atmospheric motions, while the negative wind speeds represent upward motions. In
503 addition, it is well known that atmospheric pollutant transport trajectories are deeply affected by
504 synoptic systems. As shown in the fifth graphs in Figs. 7-11, to reveal how the typical synoptic
505 weather patterns influence the distribution of particles in the YRD, the 72-h backward trajectories
506 are calculated and then clustered. Given that Nanjing is the most polluted city in the YRD, as
507 described in Section 3.1, the observational site in Nanjing (32°N, 118.8°E) is chosen for the
508 terminus of the trajectory of each synoptic weather pattern.

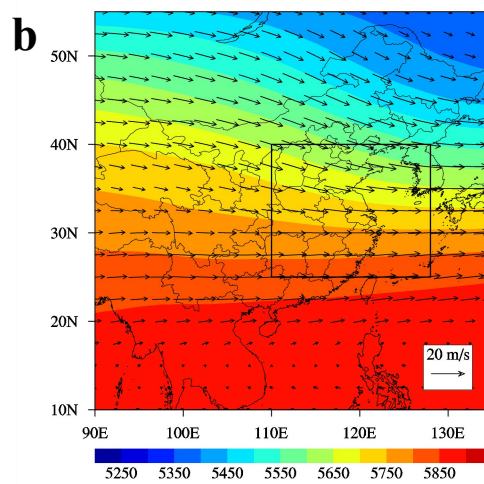
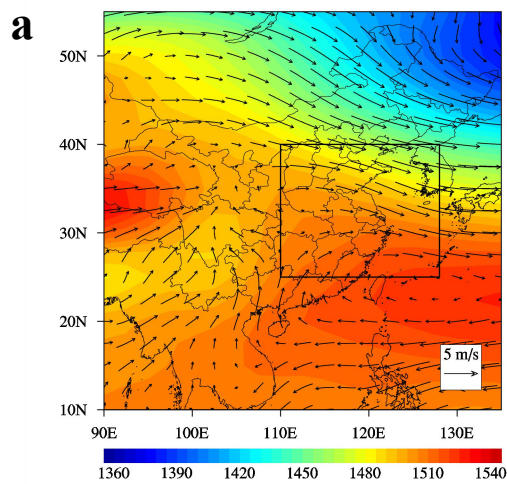
509 As illustrated in Fig. 7a, Pattern 1 usually occurs when the YRD is located at the rear of the
510 East Asian major trough and is under the control of a high-pressure ridge at 850 hPa. The center of
511 the high-pressure system is located in the northwestern Pacific Ocean. Meanwhile, northeastern
512 China is strongly affected by a low-pressure system, namely, the Aleutian Low. The strong
513 horizontal northwest wind at the rear of the East Asian major trough can transport pollutants from
514 the BTH (with high AOD, as shown in Fig. 6k) to the YRD. At the same time, the west and
515 southwest wind at the rear of the high-pressure ridge can also transport pollutants from central and
516 southwestern China (such as the SCB and Guangxi Province) to the YRD. The confluence of air
517 flows may cause an accumulation of pollutants in the YRD. Accordingly, the atmospheric
518 circulation at 500 hPa features a shallow through with a west-northwest flow (Fig. 7b). The sea
519 level pressure pattern is nearly dominated by a uniform pressure field, which exhibits relatively
520 weak anti-cyclonic circulation over the YRD (Fig. 7c). The above discussion can be further
521 explained by the 72-h backward trajectories displayed in Fig. 7e. When the YRD is under the
522 control of Pattern 1, the air masses are mainly from northern China (44%), followed by the central
523 (36%) and northeastern regions of the YRD (19%). This suggests that particle pollution is
524 remarkably affected by the polluted air masses from the BTH and the central city clusters. Surface
525 meteorological observation records also indicate that west-northwest-southwest surface winds are
526 dominant in Nanjing (Fig. 7f) and that high $\text{PM}_{2.5}$ is closely associated with the transport of
527 polluted air masses in these wind directions. In the vertical section (Fig. 7d), the relatively weak

528 upward air flows are dominant to the south of 30°N, while clear downward air flows are prevalent
 529 to the north of 30°N. The largest descending velocity ($\sim 8 \times 10^{-2} \text{ Pa s}^{-1}$) appears at an altitude of 500
 530 hPa and a latitude of 37.5°N. Downward motion is dominant above the YRD, which is in
 531 accordance with the 850 hPa circulation pattern represented by a high-pressure ridge. Thus, the
 532 weather conditions are relatively stable near the surface, which is beneficial to the local
 533 accumulation of pollutants. Overall, Pattern 1 represents a stable synoptic weather pattern that is
 534 extremely conducive to the build-up of atmospheric pollutants over the YRD. This result is
 535 consistent with the findings of Zheng et al (2015b).

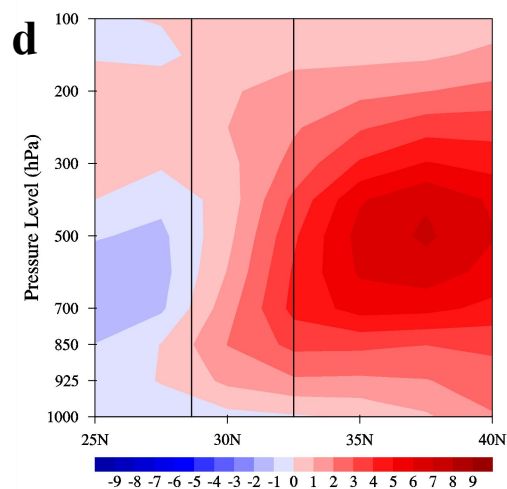
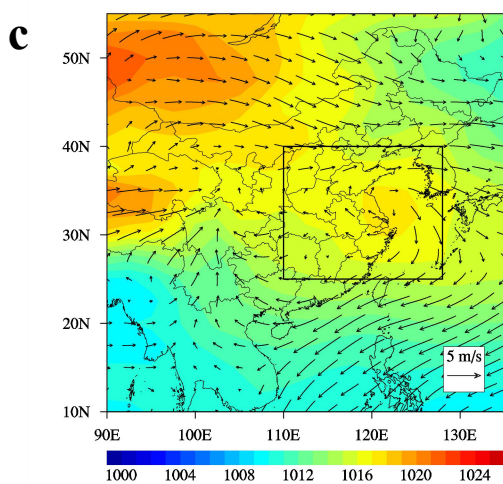
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Pattern 1

537

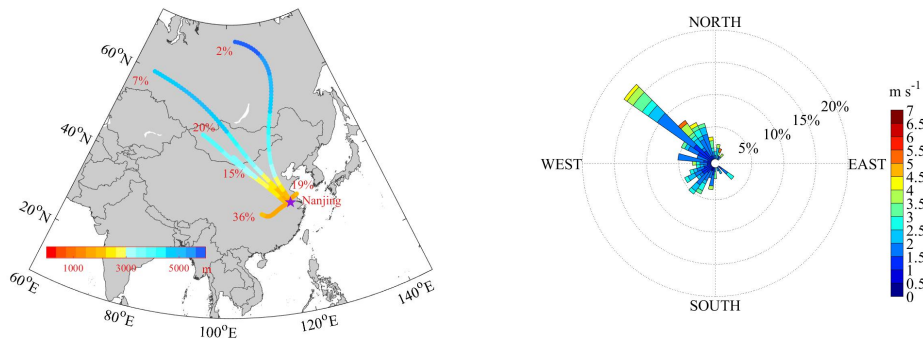


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e

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539

540

541 **Figure 7. Weather condition in Pattern 1. (a) 850 hPa geopotential height field and wind field; (b) 500 hPa**
 542 **geopotential height field and wind field; (c) sea level pressure field and 1000 hPa wind field; (d)**
 543 **height-latitude cross-sections of vertical velocity (unit: 10^{-2} Pa/s) averaged from longitude of 110-128°E; (e)**
 544 **72-h backward trajectory ending at a height of 1500 m; and (f) observation wind rose plots in Nanjing. In**
 545 **(a)-(c), the highlighted boxes denote the study area (i.e., the YRD). In (d), the black rectangular region**
 546 **represents the 16 cities in the YRD (28.6-32.5°N). In (e), the purple marker indicates the location of Nanjing**
 547 **(32°N, 118.8°E). These data represent averages for all days corresponding to Pattern 1.**

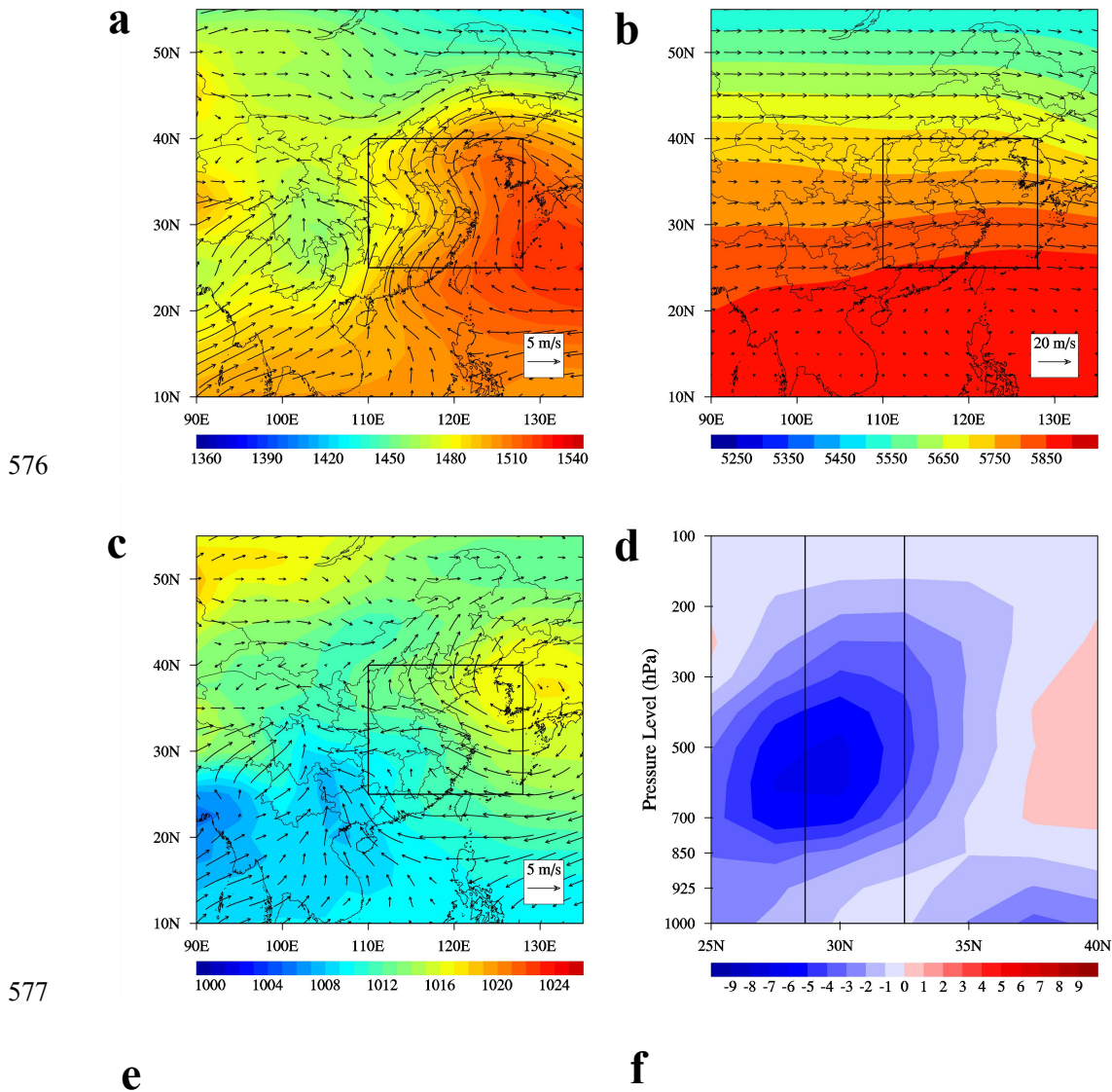
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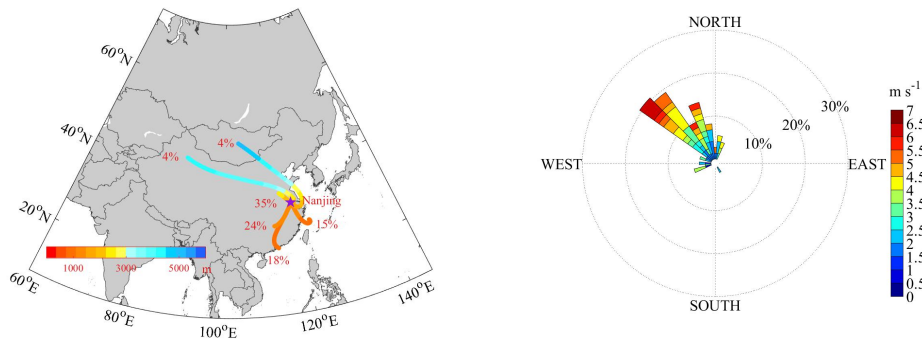
549 In Pattern 2, a low-pressure center (the Southeast Vortex) is centered in the SCB, the East
 550 China Sea is influenced by a high-pressure system, and a depression inverted trough extends and
 551 covers the YRD region at a latitude at 850 hPa (Fig. 8a). Consequently, in the YRD, the strong
 552 southwest air flows from southern China meet with the southeast air flows from the East China
 553 Sea. After the convergence of these air masses, they jointly transport pollutants northwestward. In
 554 contrast, at the surface (Fig. 8c), the study area is located at the bottom of a high-pressure system
 555 and is impacted by a strong southeast wind. In the middle troposphere (Fig. 8b), the sparse
 556 isopleths indicate that there is a small geopotential height gradient, while the shallow ridge causes
 557 westerly flows. Fig. 8e also illustrates these air pollutant transport paths. For the days when
 558 Pattern 2 is dominant, approximately 42% of the air masses are from the southwest and the south
 559 of China, and 15% are from the East China Sea. The air masses from the East China Sea are very
 560 important because the clean marine air masses may dilute the particle concentrations in the YRD.
 561 In addition, nearly 43% of air masses originate from the local sources of the YRD, which may be
 562 related to their short-range transport in the northwest direction. This is also in accordance with the
 563 dominant northwest surface wind in Nanjing (Fig. 8f). In regard to its vertical structure (Fig. 8d),
 564 Pattern 2 is obviously different than Pattern 1, as upward air flows are dominant to the south of
 565 37.5°N. The largest updraft zone ($\sim 7 \times 10^{-2}$ Pa s⁻¹) appears above the YRD and between the

566 altitudes of 700 hPa and 500 hPa. The vertical velocity close to the surface is weaker than that at
 567 higher levels over the YRD. Meanwhile, stronger upward motion occurs near the surface at a
 568 latitude of 37.5°N, with weak downward motion occurring above the 700 hPa layer. The above
 569 discussion suggests that atmospheric pollutants in the YRD are horizontally transported
 570 northwestward to a higher latitude and vertically transported upward to higher layers. Therefore,
 571 despite the transport of abundant pollutants to the YRD via southwest air flows and the
 572 short-range northwest transport of polluted air masses, the strong surface southeast wind and
 573 upward motion under the weather situation of Pattern 2 result in much less particle pollution over
 574 the YRD compared to Pattern 1.

575

Pattern 2





578

579 **Figure 8. As in Fig. 7, but for Pattern 2.**

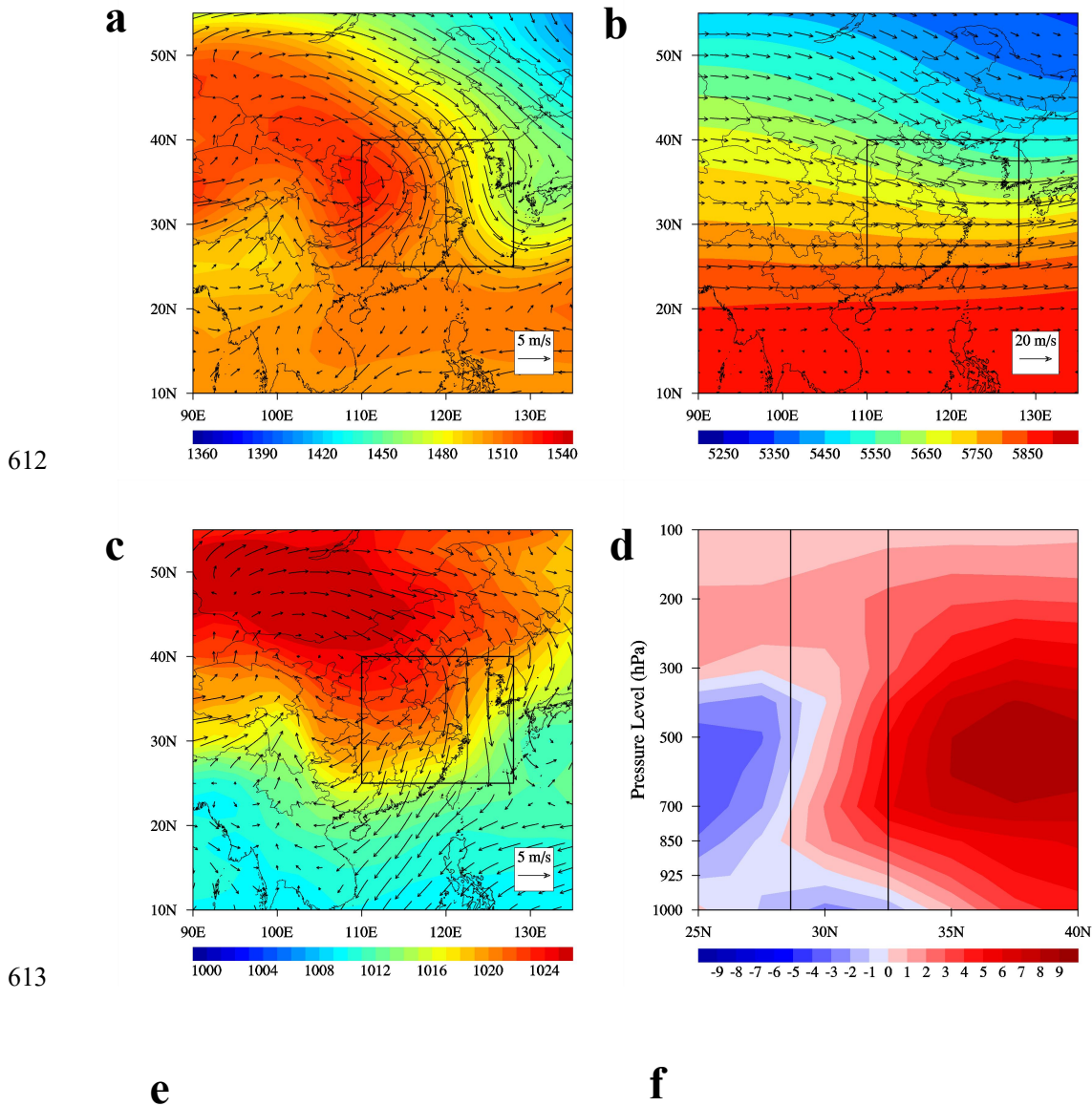
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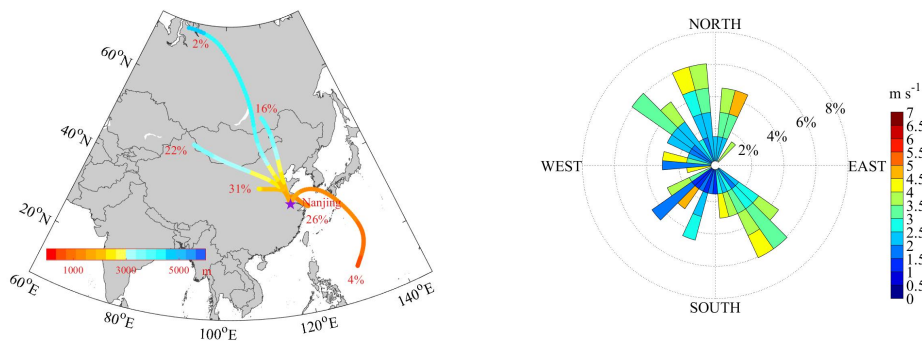
581 Pattern 3 tends to occur in winter (36.4%, as displayed in Table 3). Under this circumstance,
 582 the YRD is mainly controlled by a high-pressure system that is centered in central China (Fig. 9a).
 583 Meanwhile, northeastern China is under the steering influence of the northwest air flows at the
 584 rear of the East Asian major trough, with its trough axis appearing along the eastern coastline of
 585 China. Affected by the strong northwest winds coming from northern China, the polluted air
 586 masses from the BTH are easily transported to the YRD. At the higher layer of 500 hPa (Fig. 9b),
 587 the circulation structure patterns are similar to those of Pattern 1. A trough appears in the upper
 588 atmosphere, resulting in relatively strong west-northwest flows. The presence of dense isopleths
 589 indicates that there is a large geopotential height gradient and strong downward flows. At the
 590 surface layer (Fig. 9c), the presence of strong northerly wind is also evident, and the YRD is
 591 located at the bottom of a high-pressure system centered in the remote Mongolian region. The
 592 above discussion is further supported by the results of back trajectory calculations. As suggested
 593 in Fig. 9e, most air masses in clusters are from the Loess Plateau (31%). The transport path of this
 594 cluster is relatively short, which may be attributed to its strong anti-cyclonic circulation. Due to
 595 the strong northerly wind, the long-range transport of air masses from remote Mongolia and
 596 northern China account for 22% and 18% of all trajectories, respectively. In addition, the local
 597 transport of air masses from the southeast coastal area in the YRD accounts for 26% of all
 598 trajectories, and the marine air masses cluster that originates from the western Pacific via the
 599 Yellow Sea accounts for 4% of all trajectories. For the vertical structure (Fig. 9d), the distribution
 600 of the vertical flow field is similar to that of Pattern 1, whereas the vertical wind is slightly
 601 stronger in the weather system of Pattern 3. Due to the influence of the high-pressure system,
 602 downward air flows are dominant to the north of approximately 28°N (including the YRD) below

603 an altitude of 300 hPa. The largest descending velocity ($\sim 9 \times 10^{-2} \text{ Pa s}^{-1}$) also appears at an altitude
 604 of 500 hPa, covering the latitude of 35-40°N. However, despite the higher surface pressure (Figs.
 605 6i and 9c) and stronger downward motion (Fig. 9d), the surface wind is also much stronger for
 606 Pattern 3 (Figs. 6g, 9a and 9c), which alleviates the problems of air pollution over the YRD
 607 compared to Pattern 1. Overall, under the weather situation of Pattern 3, the strong northwest wind
 608 in the front of the high-pressure system usually leads to the transport of polluted air masses from
 609 the BTH to the YRD. Nevertheless, the strong surface wind is conducive to the mitigation of
 610 pollutants, which plays a significant role in the level of air pollution over the YRD.

611

Pattern 3





614

615 **Figure 9. As in Fig. 7, but for Pattern 3.**

616

617 In Pattern 4, on both the surface and at the 850 hPa level, the study area is under the control
 618 of a high-pressure system (Figs. 10a and 10c). The center of the high-pressure system is located in
 619 the Sea of Japan, while a cyclonic circulation occurs over the Philippine Sea. Anti-cyclonic
 620 circulation prevails over the YRD and horizontally brings the clean marine air masses to the land.
 621 Meanwhile, the sparse isopleths represent a small geopotential height gradient in the middle
 622 troposphere, which is accompanied by a much weaker west wind compared to the other patterns
 623 (Fig. 10b). Accordingly, influenced by the high-pressure system, downward atmospheric motion is
 624 clearly dominant in the vertical direction (Fig. 10d). The strongest downward motion ($\sim 6 \times 10^{-2}$ Pa
 625 s^{-1}) appears between the altitudes of 300 hPa and 500 hPa at a latitude of $35^{\circ}N$. The weak updrafts
 626 near the surface may be related to the regional thermodynamic circulation. As shown in Fig. 10e,
 627 the cluster with the largest frequency of 32% represents the local transport of air masses from the
 628 southern adjacent areas in the YRD. Additionally, the air masses originating from northern China
 629 via the Bohai Bay (25%), from Japan via the Yellow Sea (23%), and from the Philippines via the
 630 East China Sea (5%) are also representative. In total, the clusters that pass over the ocean areas
 631 account for more than 50% of all trajectories. Therefore, under this weather situation, the dilution
 632 effects of clean marine air masses play a large role in the particle pollution over the YRD.

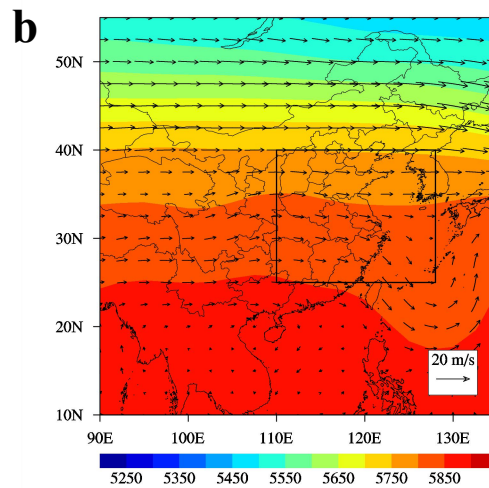
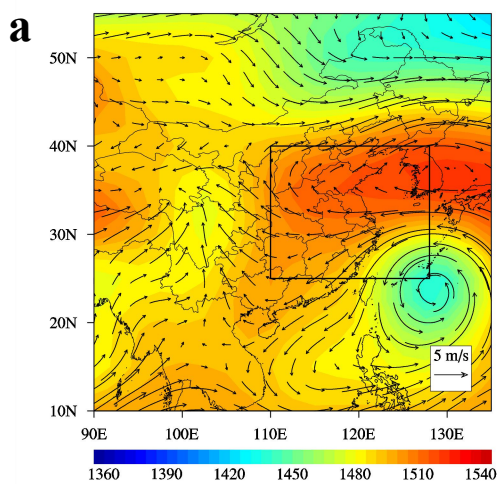
633 Pattern 5 features one of the most complex circulation situations at 850 hPa (Fig. 11a). The
 634 YRD is located between the bottom of the northern high-pressure system and the top of the
 635 southern weak low-pressure system. Thus, the strong horizontal east wind prevails and easily
 636 carries clean marine air masses from the East China Sea to the YRD. The corresponding
 637 circulation structure at the surface layer is similar to that at the 850 hPa layer (Fig. 11c), while

638 east-northeast flows are prevalent over the study domain. In the upper troposphere, a ridge appears
 639 in the east due to the tropical cyclonic system, thus leading to the west-southwest flows over the
 640 region. Due to the abovementioned two opposite pressure systems (Fig. 11a), strong upward air
 641 flows are dominant to the south of the latitude of 35 °N, while downward motion is obvious in the
 642 north (Fig. 11d). The largest ascending velocity ($\sim -9 \times 10^{-2} \text{ Pa s}^{-1}$) appears at a latitude of
 643 approximately 27.5 °N in the upper troposphere. This strong upward motion facilitates the
 644 diffusion and removal of the accumulated pollutants from the surface layer. According to Fig. 11e,
 645 the cluster with the largest frequency of 45% consists of the wet air parcels originating from Japan
 646 via the Yellow Sea. Only 5% of the trajectories originate from the Philippines and pass over the
 647 East China Sea. Overall, under the weather situation of Pattern 5, the transport of clean marine air
 648 masses and favorable diffusion conditions contribute to the good air quality over the YRD.

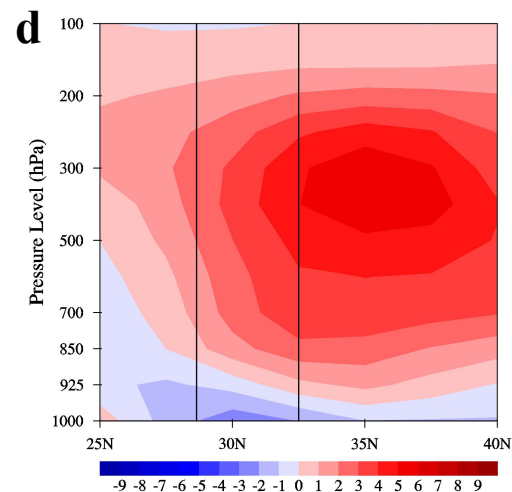
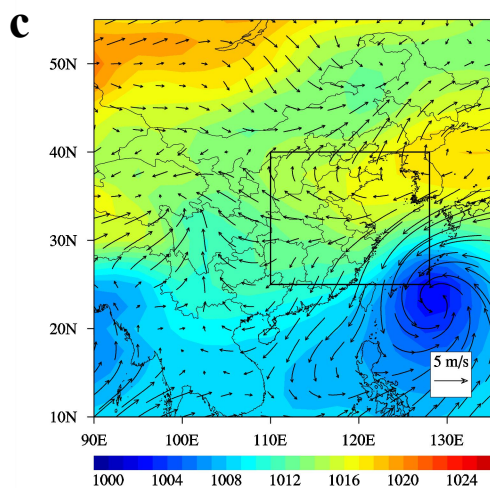
649

Pattern 4

650

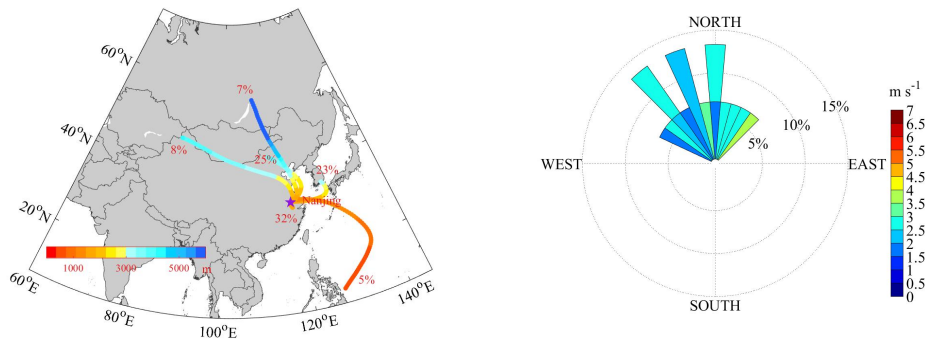


651



e

f



652

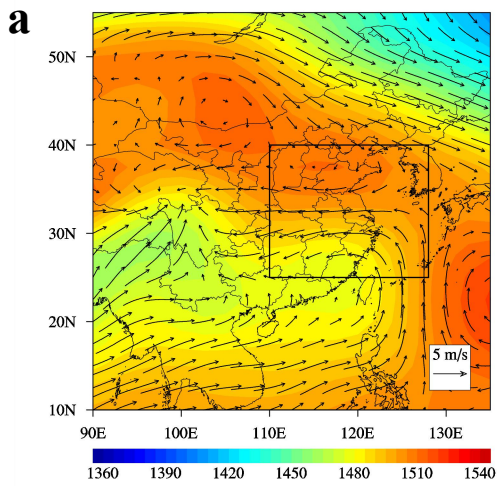
653

654 Figure 10. As in Fig. 7, but for Pattern 4.

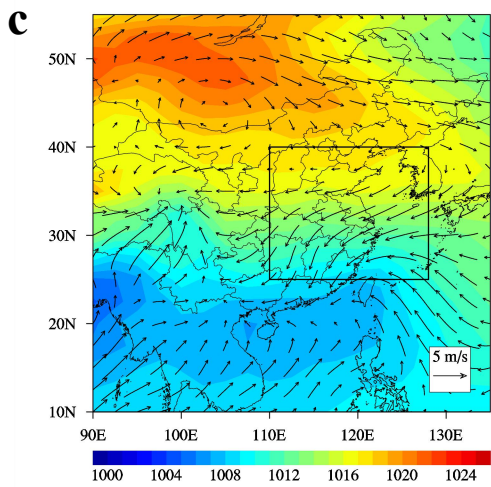
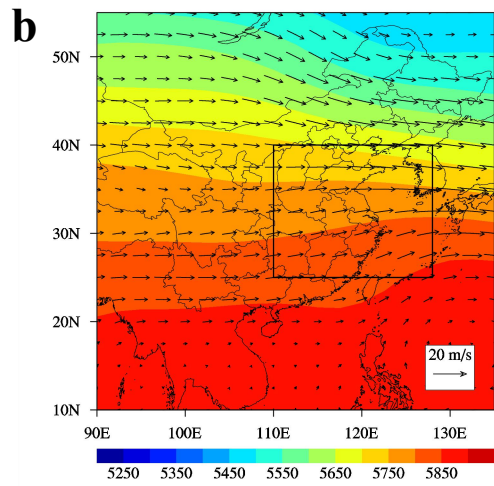
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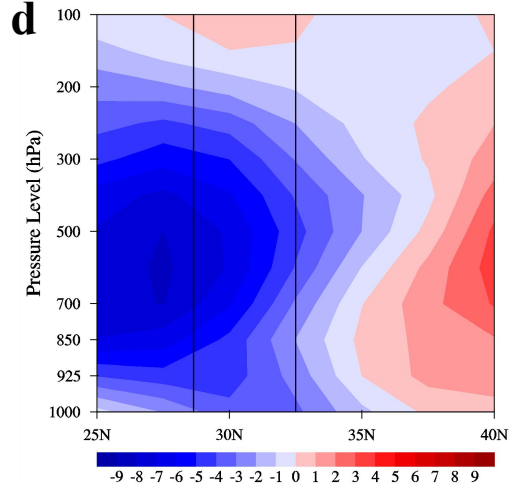
Pattern 5



657



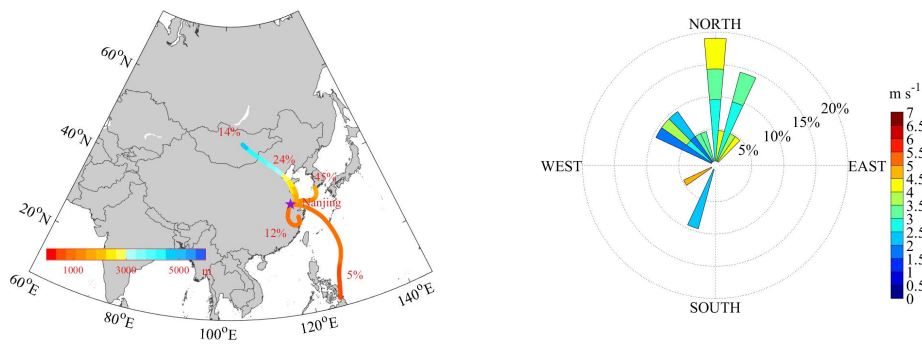
658



e

30

f



659

660 **Figure 11.** As in Fig. 7, but for Pattern 5.

661

662 To summarize, the weather situations for Patterns 1-5 are more or less affected by a
 663 high-pressure system. However, the relative positions of the study area to the anti-cyclonic
 664 circulation system have significant effects on the air quality of the YRD. These differences
 665 determine the wind speed and wind direction, and the latter further determines whether the YRD is
 666 influenced by the clean marine air masses. In both Pattern 1 and Pattern 3, the YRD is impacted
 667 by the northwest air flows at the rear of the East Asian major trough, which transport abundant air
 668 pollutants from other regions (such as the BTH and the SCB) to the YRD and cause severe particle
 669 pollution (as well as high AOD values) in the YRD. In contrast, the weaker local surface wind in
 670 Pattern 1 is extremely conducive to the local accumulation of pollutants. For this reason, Pattern 1
 671 is ‘the most polluted pattern’, and it is responsible for most of the large-scale particle pollution
 672 episodes over the YRD. Due to its stronger surface wind, Pattern 3 is ‘the second-most polluted
 673 pattern’. In Pattern 2, the polluted air masses mainly travel from the southern inland areas and
 674 synchronously meet with the clean marine air masses in the YRD. To some extent, this weather
 675 situation helps mitigate particle pollution in the YRD. In Pattern 4 and Pattern 5, the YRD is
 676 directly influenced by air flows traveling from the ocean areas, and it is thus unlikely to be
 677 polluted. Thus, Pattern 4 and Pattern 5 can be identified as ‘the clean patterns’. These data suggest
 678 that the clean marine air masses can substantially dilute the particle pollution over the YRD.

679

680 **4. Conclusions**

681 In this study, the spatial and temporal distributions of particle pollution in 16 YRD cities are
 682 characterized from December 2013 to November 2014. Meanwhile, synoptic weather
 683 classification is conducted to identify the dominant weather patterns over the YRD. The

684 meteorological fields and 72-h backward trajectories are analyzed to reveal the potential impacts
685 of weather systems on regional severe particle pollution episodes.

686 Observational records indicate that the concentrations of PM_{2.5} and PM₁₀ decrease
687 progressively in the northwest-southeast direction. The pollution levels are comparatively higher
688 in Jiangsu Province and much lower in the southeast coastal area (i.e., Ningbo, Taizhou and
689 Zhoushan). The highest particle concentrations occur in Nanjing, where the concentrations of
690 PM_{2.5} and PM₁₀ are 79 and 130 $\mu\text{g}\cdot\text{m}^{-3}$, respectively. The PM_{2.5}/PM₁₀ ratios are high in the YRD,
691 especially in winter. The seasonal mean PM_{2.5}/PM₁₀ ratios are 0.73 (winter), 0.61 (spring), 0.67
692 (summer) and 0.63 (autumn). These high PM_{2.5}/PM₁₀ ratios suggest that the PM_{2.5} fraction is
693 extraordinarily dominant in the PM₁₀ mass in the YRD. In addition, high AOD values are also
694 found in the YRD, with an annual mean value of 0.71 ± 0.57 and a maximum seasonal mean value
695 of 0.98 ± 0.83 in summer. The diurnal cycles of the particle concentrations in most cities follow the
696 same pattern, reaching a morning peak from 8:00 to 12:00. There are three peaks in seasonal
697 variations (December, March, and May or June). The wintertime peak is closely related to
698 enhanced emissions during the heating season and poor meteorological conditions. Moreover, the
699 YRD suffers from PM_{2.5} (PM₁₀) pollution on nearly 28.0% (13.1%) of the days of the year.
700 Continuous large-scale regional PM_{2.5} pollution episodes occur much more frequently than PM₁₀
701 pollution episodes.

702 Based on the sums-of-squares technique, five typical synoptic weather patterns are
703 objectively identified in the YRD, including Pattern 1 (northwestly inland wind, which occurs on
704 47.7% of all days), Pattern 2 (southwestly, 20.0%), Pattern 3 (northly inland wind, 18.1%), Pattern
705 4 (cyclone-related, 4.1%) and Pattern 5 (oceanic circulation related, 5.8%). Each pattern differs
706 from the other in respect to the relative position of the YRD to the main synoptic system (the
707 anti-cyclonic circulation system). This difference determines the wind speed and wind direction,
708 which play important roles in the air quality level of the YRD. In particular, the wind direction is
709 closely associated with determining whether the YRD is influenced by clean marine air masses. In
710 the patterns in which the YRD is located at the rear of the East Asian major trough at 850 hPa
711 (Pattern 1 and Pattern 3), strong northwest wind can easily transport air pollutants from other
712 polluted areas to the YRD, thus leading to serious particle pollution in the YRD. Due to the
713 high-pressure system, significant vertical downward motion is dominant above the YRD, resulting

714 in relatively stable weather conditions at the surface. With weak local surface wind, the worst
715 polluted weather pattern (Pattern 1) features the highest regional mean PM_{10} ($116.5 \pm 66.9 \mu\text{g}\cdot\text{m}^{-3}$),
716 $PM_{2.5}$ ($75.9 \pm 49.9 \mu\text{g}\cdot\text{m}^{-3}$) and high AOD (0.74) values. Pattern 1 is also responsible for most of the
717 large-scale regional $PM_{2.5}$ (70.4%) and PM_{10} (78.3%) pollution episodes in the YRD. In Pattern 3,
718 the strongest surface wind is conducive to the mitigation of pollution, thus resulting in the
719 second-highest PM_{10} ($86.9 \pm 49.5 \mu\text{g}\cdot\text{m}^{-3}$) and $PM_{2.5}$ ($59.1 \pm 37.3 \mu\text{g}\cdot\text{m}^{-3}$) values. In contrast, under
720 the weather system of other synoptic patterns (especially Pattern 4 and Pattern 5), the clean marine
721 air masses, which are transported via the east-southeast wind, play a crucial role in the mitigation
722 of pollution over the YRD. Therefore, the YRD has a much smaller chance of being polluted.

723 In summary, the above results reveal that particle pollution in China is a thorny issue not only
724 over a single city but also on a regional scale. This study can enhance our understanding of the
725 features of particle pollution in East Asia. Meanwhile, these results also confirm that large-scale
726 synoptic weather systems exert large impacts on regional particle pollution. Therefore,
727 establishing potential links between different levels of particle pollution and predominant synoptic
728 patterns can provide insight into formulating pollution control and mitigation strategies.

729

730 **5. Data availability**

731 The air quality monitoring records are available at <http://106.37.208.233:20035>. The
732 meteorological data are available at <http://www.nmc.cn>. The MODIS/AOD records are available
733 at <https://ladsweb.nascom.nasa.gov/search/index.html>. The NCEP reanalysis data are available at
734 <https://www.esrl.noaa.gov/psd/data/gridded/data.ncep.reanalysis2.pressure.html> and
735 <http://ready.arl.noaa.gov/archives.php>.

736

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743

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