1   **Dominant synoptic patterns associated with the decay process of**

2   **PM2.5 pollution episodes around Beijing**

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Abstract

16 The variation in the concentrations of ambient PM2.5 (particles with an aerodynamic diameter less

17 than 2.5 μm) generally forms a continuous sawtooth cycle with a recurring smooth increase followed

18 by a sharp decrease. The abrupt decay of pollution episode is mostly meteorological in origin, and

19 is controlled by the passage of synoptic systems. One affordable and effective measure for the

20 quickly reducing PM2.5 concentrations in northern China is to wait for strong wind to arrive.

21 However, it is still unclear how strong the wind needs to be and exactly what kind of synoptic system

22 most effectively results in the rapid decay of air pollution episodes. PM2.5 variations over the 28

23 pollution channel cities of Beijing are investigated to determine the mechanisms by which synoptic

24 patterns affect the decay processes of pollution episodes. This work shows more obvious day-to-

25 day variations in PM2.5 concentration in winter than in summer, which implies that wintertime

26 PM2.5 variations are more sensitive to meteorological factors. There were 365 decay processes from

27 January 2014 to March 2020, and 97 of them were related to the effective wet deposition. 26%~43%

28 of PM2.5 pollutant is removed by the wet deposition in different seasons. Two dominant circulation

29 patterns are identified in summer, and the same three circulation types (CTs) are identified in the

30 other three seasons based on the dry-day cases. The circulation patterns beneficial to the decay

31 processes all exhibit a higher than normal surface wind speed, a negative relative humidity anomaly

32 and positive divergence in the PM2.5 horizontal flux. In addition, CT1 in spring, autumn and winter

33 is controlled by northeasterly wind and features the most significant horizontal net-outflow of air

34 pollutants and effective upward spread of air pollutants to the free atmosphere, which promotes the

35 abrupt reduction of local PM2.5 concentrations. CT2 is the most frequent synoptic pattern leading

36 to decay processes in autumn and winter, and the domain region is located to the east of an

37 anticyclone system. CT2 features a strong northwesterly wind of 2.98~3.88 m/s, the lowest relative

38 humidity and the highest boundary layer height (BLH) among the three CTs, all of which are

39 favorable for the reduction of PM2.5 concentrations. In CT3, a prevailing westerly wind anomaly

40 occurs in the domain, with remarkable zonal divergence in the PM2.5 flux and strong horizontal

41 wind shear in the near-surface under the boundary layer. PM2.5 concentrations show significant

42 decreases of more than 37%, 41% and 27% after the passage of CT1, CT2 and CT3, respectively. A

43 dry air mass with a positive BLH anomaly and the effective horizontal outflow of air pollutants are

44 the main reasons for the abrupt decay phase in summer. PM2.5 concentrations after the decay

45 process show a significant decreasing trend from 2014 to 2020, reflecting successful emission

46 mitigation. Emission reductions have led to a 4.3~5.7 μg/(m^3.yr) decrease in PM2.5 concentrations

47 in the 28 pollution channel cities of Beijing.

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1. Introduction

PM2.5 pollution (particles with an aerodynamic diameter less than 2.5 μm) has become a severe threat and challenge in China, especially in the Beijing-Tianjin-Hebei (BTH) region, and has attracted significant concern regarding how to improve regional air quality (Che et al., 2019;Wang et al., 2019a;Xia et al., 2016;Zhang et al., 2018;Mu and Zhang, 2014;Cai et al., 2017;Wang et al., 2015). To avoid the severe negative impacts of air pollution on public health, the Chinese government has issued a number of policies to improve the atmospheric environment (Ding et al., 2019;Chen and Wang, 2015;Zhao et al., 2019;Li et al., 2018b). For example, in September 2013, the State Council issued the Air Pollution Prevention and Control Action Plan (referred to as Clean Air Action), which required the BTH region to reduce its PM2.5 concentrations by 25% within 5 years (China's State Council, 2013). With the deep research on the prevention and control of air pollution, the regional effects of air pollution from cities in the pollution transmission channel in the BTH region have been highlighted (China Daily, 2017). Therefore, the Work Plan for Air Pollution Prevention and Control in Beijing, Tianjin, and Hebei and Surrounding Areas was released in March 2017 (China's State Council, 2018). Much stricter, more comprehensive, and more detailed prevention and control measurements were taken in the "2+26" cities, including Beijing, Tianjin, and 26 other cities in the provinces of Hebei, Shandong, Henan and Shanxi. Due to the persistent efforts towards emission mitigation, the air quality has shown significant improvement in these 28 pollution channel cities in recent years (Zhang et al., 2019a;Zhang et al., 2019b;Zheng et al., 2018;Wang et al., 2019d;Gui et al., 2020).

Meteorological conditions are considered as one of the important factors for the variation in ambient PM2.5 pollution, especially for the temporal evolution of each air pollution episode (Zhang et al., 2014;Ma and Zhang, 2020;Wang et al., 2019c). Even under the conditions of a significant decrease in air pollutant emissions, similar to the COVID-19 lockdown period, PM2.5 pollution events still occur frequently in the 28 pollution channel cities due to the unfavorable meteorological background (Shi and Brasseur, 2020;Le et al., 2020;Huang et al., 2020b;Wang et al., 2020b; Wang and Zhang, 2020b). Many studies have been conducted and have suggested that multiple meteorological factors influence the emission of primary pollutants, the formation of secondary particles and the processes of transport, accumulation and deposition of particles (Zhao et al., 2020a;Huang et al., 2020c;Chen et al., 2019;Gong and Liao, 2019). High temperatures result in greater emissions of PM2.5 precursors and secondary pollutants, and promotes photochemical reactions, causing an increase in local PM2.5 concentrations (Zhang, 2017;Zhao et al., 2018b;Chen et al., 2020). Humidity strongly affects PM2.5 concentrations in China, especially during severe pollution episodes (Zhao et al., 2018a;Li et al., 2018a;Huang et al., 2020a). Higher humidity is

83   beneficial for the hygroscopic increase in aerosols and facilitates the formation of secondary

84   particles (Wang et al., 2019b;Zhao et al., 2017;Cheng et al., 2015;Xin et al., 2016). The cross-

85   regional transport and horizontal diffusion of pollutants are strongly determined by the wind field.

86   Southerly winds bring higher concentrations of air pollutants and more moisture, which enhances

87   the local air pollution in Beijing and the surrounding regions (He et al., 2020;Zhao et al., 2020b). In

88   addition to individual meteorological variables, synoptic circulation characteristics control the

89   formation and development of air pollution events (Wang et al., 2020a;Miao et al., 2020;Wang and

90   Zhang, 2020a; Liu et al., 2019). Monsoonal flows and cold frontal passages are the dominant

91   meteorological modes controlling the day-to-day variations in PM2.5 concentrations in the northern

92   China (Li et al., 2016;Wu et al., 2017;Zhang et al., 1996;Leung et al., 2018). Weak synoptic patterns

93   with high-pressure or persistent low-pressure systems favor the accumulation of pollutants, while,

94   strong synoptic patterns with large pressure gradients encourage the diffusion of pollutants (Cai et

95   al., 2020;Zhang et al., 2017;Zhang et al., 2020;Li et al., 2019). Severe haze events in the BTH region

96   are always accompanied by stagnant air conditions, stable stratification, weak surface wind, low

97   boundary layer height (BLH), and high relative humidity (Ma et al., 2020;Bi et al., 2014;Wang et

98   al., 2020c;Tang et al., 2016;Quan et al., 2020;Pei et al., 2020;Guo et al., 2019).

99   Most of the aforementioned studies focused on the synoptic pattern characteristics favorable for the

100   initiation and development of air pollution episodes in the BTH region. During the developing phase

101   of each PM2.5 pollution episode, the comprehensive effects of secondary aerosol formation,

102   hygroscopic increase and accumulation of particles lead to an increase in local PM2.5

103   concentrations, which usually takes several days from a clean situation to the outbreak of a heavy

104   haze (Sun et al., 2014;Wang et al., 2016;Pei et al., 2018). Both atmospheric chemistry and physics

105   processes play important roles in the developing phase of air pollution events (Gu et al., 2020;Yao

106   et al., 2018;Wang et al., 2018;Wang et al., 2010;Li et al., 2017;Gao et al., 2017). However, compared

107   to the developing phase, which typically features a smooth increase in air pollutant concentrations,

108   the decay phase of each pollution episode shows a sharp decrease in PM2.5 concentrations, often in

109   a few hours. The abrupt decrease in PM2.5 concentrations is purely meteorological in origin and is

110   controlled by the passage of synoptic systems, especially cold fronts, which terminate a severe air

111   pollution episode in the BTH region by strong winds (Zhu et al., 2016;Jia et al., 2008;Ji et al.,

112   2012;Xin et al., 2012). However, it is still unclear how strong the wind needs to be, exactly what

113   kind of synoptic systems can effectively terminate air pollution episodes in the BTH region, and

114   what mechanism is responsible for the rapid reduction in PM2.5 concentrations in a few hours. The

115   clarification of these issues will contribute to improving local air quality predictions. The variation

116   in air quality is generally consistent in the 28 pollution channel cities, especially in the decay phase

117   of pollution episodes, which indicates that the same synoptic system usually affects the whole region.

118   This study will focus on the region covering these 28 pollution channel cities and reveal the synoptic

119   circulation pattern that dominates the decay process of PM2.5 pollution events.

2. Data and Method

2.1 Dataset

122   The daily mean observed PM2.5 concentrations in the 28 pollution channel cities from January 2014

123   to March 2020 were obtained from the Ministry of Ecology and Environment of the People's

124   Republic of China (https://www.aqistudy.cn/historydata/). Fig. 1 shows the location of the 28

125   pollution channel cities and their annual mean PM2.5 concentrations from 2014 to 2019. The four-

126   times-daily dataset of the fifth-generation European Centre for Medium-Range Weather Forecasts

127   (ECMWF   ERA5)   atmospheric   reanalysis   dataset   with   a   resolution   of   0.5°

128   (https://cds.climate.copernicus.eu/cdsapp#!/dataset/10.24381/cds.bd0915c6?tab=form) was used to

129   describe the meteorological characteristics and synoptic circulation classification.

130   The divergence of local PM2.5 flux can be taken as a metric for the PM2.5 budget in the specific

131   region, with positive divergence indicating net outflow of air pollutants from the domain region,

132   and vice versa. The daily mean divergence of the PM2.5 flux over the region of 34-40° N and 112-

133   118° E is calculated according to Eq.(1):

134    $$D = D_Z + D_m = \frac{\partial}{\partial x}(UQ) + \frac{\partial}{\partial y}(VQ) = \sum_{i=1}^{n} \frac{(U_{Ei}Q_{Ei} - U_{Wi}Q_{Wi})}{2\Delta X} + \sum_{j=1}^{m} \frac{(V_{Nj}Q_{Nj} - V_{Sj}Q_{Sj})}{2\Delta Y} \quad (1)$$

135   where D_z and D_m are the zonal and meridional components of the net divergence of PM2.5 flux for

136   the specific region. The parameters n and m indicate the meridional and zonal grid numbers of the

137   domain. The subscripts E and W mark the variables at the longitudes of the eastern and western

138   boundaries of the domain. Similarly, the subscripts S and N represent the values at the latitudes of

139   the southern and northern boundaries. U_{Ei} (units in m/s) indicates the 10 m zonal wind in the ith grid

140   of the eastern boundary of the domain. Q_{Nj} (units in μg/m^3) is the spatially interpolated PM2.5

141   concentration in the jth grid at the latitude of the northern boundary. ΔX and ΔY represent the zonal

142   and meridional distance of each grid (units in meters). Due to the limited information on the vertical

143   distribution of PM2.5 and the horizontal winds are closely related with PM2.5 concentration as

144   revealed by previous studies, the horizontal divergence of PM2.5 flux is used to evaluate the net

145   inflow and outflow of local air pollutants in this study.

2.2 Thresholds for the decay process of air pollution episodes

147   Fig. 2 shows the daily PM2.5 concentration variations of the 28 pollution channel cities from

148   January to March 2019. PM2.5 concentrations exhibit a recurring smooth increase followed by a

149   sharp decrease, which is known as a sawtooth cycle (Jia et al., 2008). During the developing phase

150   of each pollution episode, the PM2.5 concentrations show the same smoothly increasing trend with

151   slight differences in the rate of increase and magnitude in the 28 pollution channel cities (i.e., an

152   average increase trend of $10.37 \pm 42.2\ \mu g/(m^3 \cdot day)$ during January to March 2019). The

153   inhomogeneity of the PM2.5 concentration increase in the 28 cities, indicating by the large standard

154   deviation of increase trends (approximate four times the magnitude of increase trend), may be due

155   to the complicated physiochemical processes of haze formation. By contrast, as shown by dotted

156   lines in Fig. 2, regional synchronous decreases in PM2.5 concentrations occur in the decay phase of

157   pollution episodes with an average trend of $-50.06 \pm 46.83\ \mu g/(m^3 \cdot day)$. Most of the

158   consistent improvements in air quality in the decay phase can be attributed to the effects of the

159   synoptic system. Therefore, in this study, if more than 40% of the 28 pollution channel cities with

160   the day-to-day PM2.5 concentrations decreased by 30% (relative to the value of the previous day)

161   or more than 60% of the channel cities with PM2.5 concentrations decreased by 30% in two

162   successive days, it can be defined as the occurrence of the decay phase of pollution episodes. If two

163   consecutive days were defined as the decay phase, only the first day was selected be vailed and

164   retained. In total, 365 days are identified as the decay phase of pollution episodes from January

165   2015 to March 2020 (see Fig. 4) and are used for the synoptic pattern classification.

166   **2.3 Method of synoptic circulation classification**

167   The T-mode principal component analysis (PCA) method was used to objectively classify the type

168   of synoptic system dominating the decay phase of pollution episodes, as this method has an

169   outstanding performance in terms of the reproduction of predefined types and temporal-spatial

170   stabilities (Huth et al., 2008;Cavazos, 2000;Tie et al., 2015;Valverde et al., 2015;Xu et al., 2016).

171   The T-mode PCA has been widely used to investigate the general circulation patterns, climate

172   change and air quality and has been incorporated into the European Cooperation in Science and

173   Technology (COST) plan 733 toolbox (COST733: http://cost733.geo.uni-augsburg.de/cost733wiki)

174   (Philipp et al., 2014). The daily mean geopotential height (Z), U and V components at 925 hPa on

175   the 365 decay phase days are used for synoptic pattern classification. To exclude the effects of

176   seasonal variation on atmospheric circulation and to ensure that different synoptic patterns in the

177   same season are comparable, the T-mode PCA method is applied to the four seasons respectively.

178   The target region is 32-44° N and 110-122° E, as shown in Fig. 1. For each season, the three input

179   data matrixes (U, V and Z) have temporal and spatial dimensions, with spatial grids and time series

180   represented by rows and columns, respectively. To speed up computations of the T-mode PCA in

181   the COST733 toolbox, each matrix is first divided into 10 subsets. Then, the principal components

182   (PCs) are determined using the singular value decomposition for each subset, and an oblique

183   rotation is applied to the PCs to achieve better classification effects. The 10 classifications based on

184   the subsets are evaluated by the chi-square test and the subset with the highest sum is selected and

185   assigned to a type.

186   3. Results

187   3.1 Identification of the occurrence of the decay process of air pollution episodes

188   The magnitude of the day-to-day variation in PM2.5 concentrations is an important metric for

189   recognizing the occurrence of the decay phase of air pollution. Fig. 3 shows the frequency of the

190   relative day-to-day PM2.5 concentration differences in the 28 pollution channel cities during the

191   period of January 2014 to March 2020. Table 1 summarizes the occurrence frequency of the day-

192   to-day PM2.5 differences in the specific segment. It shows that a fatter-tailed probability distribution

193   exists in winter than in summer; thus, winter features a lower probability of weak PM2.5 variations

194   and a higher probability of strong PM2.5 variations, indicating greater day-to-day variability in

195   PM2.5 concentrations. In winter, 8.6% of PM2.5 concentrations decreased by over 60%, and 14.9%

196   increased by more than 80%, whereas, in summer, the values were only 2.4% and 6.6%. A total of

197   38.3% of the cases show day-to-day PM2.5 variations within the range of -20% to 40% in winter,

198   but where a total of 55.6% is observed in summer. The PM2.5 variations in spring and autumn

199   exhibit almost the same distribution patterns, with a relatively higher frequency of strong PM2.5

200   variations in autumn. Generally, the probability distributions in spring and autumn are between

201   those of summer and winter. The stronger day-to-day decreases in PM2.5 concentrations,

202   particularly the sharp wintertime reductions, may be attributable to the passage of a cold front

203   synoptic system, and the results suggest that the winter PM2.5 variations are the most sensitive to

204   synoptic patterns.

205   According to the occurrence of day-to-day PM2.5 differences in the 28 pollution channel cities, i.e.,

206   thresholds for the decay phase of air pollution episodes in Section 2.2, 365 decay processes have

207   been recognized from January 2014 to March 2020. 97 of the 365 decay phases have effective

208   precipitation more than of 10 mm/day, in which case the abrupt decrease in ambient PM2.5

209   concentrations are assumed to be related to wet deposition. Only the decay processes on dry days

210   are involved in the synoptic pattern classification in the following work. Figure 4 shows the annual

211   cycle of the decay process frequencies in the specific year. In most years, the figure shows a two-

212   peak annual cycle of the decay phase frequency with a valley in summer, and the valley becomes

213   deeper after removing the rainy cases. There are 105 (105), 62 (21), 86 (56) and 112 (109) decay

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214 process days in spring, summer, autumn and winter for all (dry-day) cases, respectively.

215 Approximately 70% of the regional sharp reduction in summer can be attributed to the effect of wet

216 deposition.

217 **3.2 Classification of the synoptic circulation dominating the decay processes of air pollution**

218 **episodes**

219 T-mode PCA circulation classification has been applied to the dry-day decay process in individual

220 seasons. Figs. 5 and 6 show the original and anomalous circulation patterns at 925 hPa under each

221 circulation type (CT) condition. Two dominant circulation types (CTs) are identified in summer, and

222 three CTs are identified in the other seasons. The three dominant CTs in spring show almost the

223 same pattern as those of autumn and winter, and only the occurrence frequency of the CTs differ

224 among the seasons. The strong prevailing northwesterly wind in the CT2 condition is the commonly

225 accepted synoptic circulation favorable for the rapid decay of pollution episodes in the BTH region,

226 and CT2 is also the most frequent CT for the decay phase in autumn and winter. A large-scale high-

227 pressure system covers the region of central-western Mongolia, northern Xinjiang, Inner Mongolia

228 and Shaanxi Province in China. Deep low pressure is situated in the northeastern China and northern

229 Japan. The BTH region is located between the east of the anticyclone and west of the cyclone, and

230 is dominated by strong northwesterly surface winds with the speeds of 2.98~3.88 m/s in different

231 seasons. The northwesterly wind corresponds to the significant northerly wind anomaly, which is

232 beneficial for the transport of cold, clean and dry air masses southward. Although it shows

233 downward motion due to the upper westerly wind passing the leeward side (see Fig. 7), the other

234 meteorological variables summarized in Fig. 8 reveal that the highest wind speed, the highest

235 boundary layer height (BLH) and the lowest relative humidity occur under CT2 conditions, all of

236 which are favorable for the reduction of PM2.5 concentrations. Fig. 9 exhibits the distribution of

237 PM2.5 flux divergence over the region of 34-40° N and 112-118° E, and its zonal and meridional

238 components, with positive divergence indicating net horizontal outflow of air pollutants from the

239 BTH region, and negative divergence indicating the opposite. The PM2.5 flux divergence is found

240 to have positive values in all three CTs, indicating that the local ambient PM2.5 concentrations

241 decrease with the removal of the polluted air mass or the replacement with clean air. The positive

242 divergence of the PM2.5 flux in CT2 is mainly contributed by the meridional outflow, which

243 highlights the effects of the northerly wind anomaly. Clean, dry and strong northwesterly winds in

244 the CT2 condition are the major drivers of the decay process of air pollution episodes.

245 In CT1 in spring, autumn and winter, a surface high-pressure system initiates from the Siberian

246 region and slants forward to central Inner Mongolia and the BTH region, resulting in a position that

247 is more northeastward than the anticyclonic circulation in CT2. Most areas in China are controlled

248   by a high-pressure system. The BTH region is located within on southeastern edge of the high-

249   pressure center with an anticyclonic horizontal wind shear in the domain. The average surface wind

250   speed is of 2.63~3.02 m/s, which is higher than the seasonal mean but not as high as that under CT2

251   conditions. Although all the surface wind speed, BLH and relative humidity show favorable patterns

252   for air pollutant diffusion under CT1 conditions, the magnitudes of the above anomalies are not as

253   high as those under CT2 conditions. Therefore, there must be other mechanisms responsible for the

254   decay process of pollution episode that are distinct from those of CT2, as is generally believed.

255   According to the anomaly pattern in Fig. 6, the BTH region is located at the south of the anticyclone,

256   which is dominated by a remarkable northeasterly wind anomaly. This northeasterly wind anomaly

257   brings clean and dry air masses to the BTH region, and increases the outward and southward

258   transport of local air pollutants in the meanwhile, which results in the negative relative humidity

259   anomaly shown in Fig. 8. The net divergence of air pollutants (i.e., positive divergence of the PM2.5

260   flux in Fig. 9) is the most significant under CT1 conditions, indicating the contribution of horizontal

261   transport to the rapid decay of pollution episodes. In terms of vertical anomaly circulation, the BTH

262   region is located under the east of a high-level ridge and west of a high-level trough (figure not

263   shown here), where there is often upper level convergence and cause the surface high-pressure

264   anomaly to get higher (see Fig. 6). The upper level convergence leads to the vertical sinking in the

265   east of the BTH region, which also delivers upper dry and clean air to the surface. In addition, as

266   shown in Fig. 7, the significant clean vertical sinking airflow in the east of the BTH region combined

267   with the surface easterly wind anomaly results in air movement westward across the domain and

268   climbs up along the western mountain region. The upward flow carries the near-surface air

269   pollutants to the upper level of the boundary layer, where the pollution quickly spreads to the free

270   atmosphere due to the effective entrainment caused by the strong wind shear at the top of the

271   boundary layer (see Fig. 7). In general, the remarkable horizontal net-outflow of air pollutants,

272   negative humidity anomaly and effective outward spread of air pollutants to the free atmosphere

273   promote the abrupt reduction of local PM2.5 concentrations.

274   CT3 is the dominant synoptic pattern for the decay process in spring, with the highest frequency of

275   47%, compared with frequencies of 30% and 17% in autumn and winter. In this kind of circulation

276   pattern, there is only a closed low-pressure system located over the northeastern China, with large

277   pressure gradients around the cyclone and weak gradients over most parts of China. The BTH region

278   borders the cyclone system to the northeast, which leads to a prevailing westerly wind with speeds

279   of 2.29~3.07 m/s. The low-pressure and westerly wind features are more significant based on the

280   anomalous circulation at 925 hPa in Fig. 6, especially in winter. In the upper 500 hPa, a deep trough

281   persists in the northern BTH region, bringing cold air masses from the northwest. Similar to CT1

282   and CT2, negative relative humidity anomalies and positive surface wind speed anomalies are

283 favorable for the decay of pollution episodes. Given the distribution of the BLH, there is no
284 significant positive anomaly signal in CT3, unlike in CT1 and CT2. Although a moderate BLH is
285 observed under CT3 conditions, strong vertical wind shear occurs near the surface, as shown in Fig.
286 7, which improves the more uniform vertical distribution of air pollutants in the boundary layer.
287 Moreover, obvious horizontal PM2.5 divergence also provides a possibility for the decay of air
288 pollution episodes. To be more precise, the zonal divergence of the PM2.5 flux that dominates the
289 net divergence of the whole region, rather than the meridional component as the other two
290 circulation patterns. The inflow of clean and dry air masses combined with the good performance
291 of boundary layer mixing are the main reasons for the immediate improvement of air quality when
292 CT3 occurs.

293 In terms of the synoptic patterns in summer, two CTs are classified excluding the effects of wet
294 deposition. According to the circulation anomaly in Fig. 6, the synoptic pattern of CT1 in summer
295 is similar to that of CT3 at 925 hPa in other seasons, which is dominated by a northeastern cyclonic
296 circulation. Dry northwesterly wind occurs in the BTH region, reducing the local relative humidity.
297 As shown in Fig. 8, the BLH is higher than the seasonal average, indicating an increase in vertical
298 diffusion space. The zonal positive divergence of the PM2.5 flux is offset by the negative value in
299 the meridional direction. The effect of horizontal transport of air pollutants can be ignored in this
300 situation. Therefore, the decay process of the air pollution episode in the CT1 condition can be
301 attributed to the dry air mass and higher than normal BLH.

302 In the anomaly pattern of the CT2 condition in summer, the BTH region is located between the
303 southern portion of a high-pressure system and the northern portion of a low-pressure system, and
304 is affected by the prevailing northeasterly surface wind. Clean air masses are transported to the BTH
305 region along with the northeasterly wind, which can be confirmed by the positive divergence in the
306 PM2.5 flux in both zonal and meridional directions. Both the negative relative humidity and positive
307 BLH anomalies in CT2 are beneficial for the reduction of surface PM2.5 concentrations, but the
308 magnitude of the anomaly is not as high as those of the CT1 condition. There is no favorable signal
309 for the diffusion of surface PM2.5 in terms of the vertical motion in the two synoptic patterns in
310 summer. It is the effective horizontal outflow that promotes the decay process of pollution episodes.

311 **3.3 Synoptic circulation effects on the PM2.5 pollution**

312 Section 3.2 shows different physical mechanisms for the rapid decay of air pollution episodes in the
313 region covering the 28 pollution channel cities. Fig. 10 exhibits the relative difference in PM2.5
314 concentrations between the day before and after the occurrence of the specific synoptic CTs. The
315 average PM2.5 differences in the 28 pollution channel cities are summarized in Table 2.
316 Unsurprisingly, it shows a remarkable decrease in PM2.5 concentrations when all the circulation

317   patterns dominate the decay process occurs, but it is worth noting that the magnitudes of the decline

318   vary according to the synoptic patterns. For the case of spring, autumn and winter, CT2 conditions

319   demonstrate the most significant effects on the abrupt reduction in PM2.5 concentrations, with a

320   more than 40% day-to-day decrease in PM2.5 concentrations in the 28 pollution channel cities in

321   all three seasons. CT1 conditions are second in terms of the circulation influence in the decay

322   process of PM2.5 pollution episodes. The PM2.5 concentrations decrease quickly by 37.2%, 40.1%

323   and 36.9% when CT1 conditions occur in spring, autumn and winter, respectively. The CT3

324   conditions, which are dominated by westerly winds, show a relatively weak ability on control the

325   decay process of PM2.5 pollution episodes. Air quality improves by approximately 26~29%

326   compared with the previous day due to the occurrence of CT3 conditions. In summer, PM2.5

327   concentrations decrease more significantly with the occurrence of CT1 conditions than with the

328   occurrence of CT2 conditions, indicating more effective diffusion under northwesterly winds than

329   under northeasterly airflow. Wet scavenging is an effective method for the rapid decay of air

330   pollution episodes, especially in wintertime. PM2.5 concentrations drop sharply after the occurrence

331   of precipitation, with decreases of more than 35% in spring, autumn and winter. 26.2% of PM2.5

332   pollution is removed by the wet deposition in summer, which is the lowest rate among the four

333   seasons. The relatively clean background may account for the weak wet deposition effects in

334   summer.

335   Fig. 2 shows the sawtooth cycle variation in PM2.5 concentrations with a smooth increase followed

336   by an abrupt decrease. However, the PM2.5 concentrations do not always increase gradually before

337   the decay of the pollution episode. Here, the sawtooth cycle is divided into developing and decay

338   phases, and the interval stage between two decay phases is defined as the developing phase of a

339   specific pollution episode. As shown in Fig. 11, when the duration of the developing phase is less

340   than 3-days, air pollutants accumulate gradually to a maximum until the occurrence of decay process

341   occurs. However, if the developing phase is longer than 3-days, the highest PM2.5 concentrations

342   occur on 1-3 days before the passage of a favorable synoptic system, which indicates that the

343   developing mature stage of pollution episodes (with high level concentrations) usually persist for

344   several days.

345   The duration of the developing phase not only changes the shape of the sawtooth cycle but also

346   affects the maximum PM2.5 concentrations during the pollution episode, as shown in Fig. 12. Most

347   of the durations of the developing phase are concentrated in the period of shorter than 5-days in

348   spring, autumn and winter, with average durations of 5.53, 5.86 and 5.36 days, respectively.

349   Typically, for the cases in spring and autumn, when the durations are less than 5 days, the maximum

350   PM2.5 concentrations during the specific air pollution episode increase with an increase in the

351   developing phase durations; but the concentrations remain unchanged if the duration longer than 5

11

352   days. In winter, the maximum PM2.5 concentrations in a specific sawtooth cycle continue to

353   increase with increases in the interval between two decay processes. Wintertime air pollution can

354   be exacerbated by the long-term absence of an effective decay process. The frequency of favorable

355   circulation patterns is relatively lower in summer, which leads to an effective decay process

356   occurring every 7.45 days. The maximum PM2.5 concentrations display an upward tendency with

357   increases in the developing stage durations, but there are some small fluctuations in the mean value

358   of the highest PM2.5 concentration due to the limited samples in summer.

359   Emission and meteorological elements are taken as the two most important factors controlling the

360   variation in PM2.5. Many efforts have been made to mitigate the air pollutant emissions in the 28

361   pollution channel cities, which have achieved remarkable improvements in air quality in recent

362   years. However, because obvious interannual difference of the meteorological conditions are

363   observed, there is uncertainty in the evaluation of emission reductions based on the observed PM2.5

364   concentrations. The quantitative evaluation of the effects of emission reduction measures on the

365   PM2.5 concentration variation has been a challenge for policy makers and stakeholders. Here, only

366   the PM2.5 concentrations observed on the days of decay processes are compared, which excludes

367   the different effects of meteorological conditions and evaluates the pure effects of emission

368   reduction from a certain perspective. Fig. 13 shows a significant decline in seasonal mean PM2.5

369   concentrations from 2014 to 2020 in the 28 pollution channel cities. This figure also shows almost

370   the same rates of decrease in all four seasons, with relatively smaller decreases of 4.8 and 4.3

371    $\mu g/(m^3 \cdot yr)$ in spring and winter and greater decreases of 5.7 and 5.2 $\mu g/(m^3 \cdot yr)$ in summer and

372   autumn, respectively. The slight difference in the seasonal decreasing tendency is possibly due to

373   difference in the main sources of air pollutant emissions.

4. Conclusions and Discussion

375   The variation in ambient air pollutant concentrations generally represents a continuous sawtooth

376   cycle with a recurring smooth increase followed by a sharp decrease. The combined effects of

377   emissions, secondary formation of particles and unfavorable meteorological conditions that trigger

378   the initiation and development of the specific PM2.5 pollution episodes over several days. In

379   contrast, the abrupt decay of pollution episodes is mostly due to the passage of favorable synoptic

380   patterns, and it usually takes a few hours for conditions to transition from hazy to clean air condition.

381   The detailed atmospheric circulation features and the mechanisms through which they affect the

382   decay processes of pollution episodes are discussed in this work. A relatively fatter-tailed probability

383   distribution of day-to-day PM2.5 concentration over the 28 pollution channel cities is observed in

384   winter compared with summer, indicating that winter features a lower probability of weak PM2.5

385   variations and a higher probability of strong PM2.5 variations. The probability distribution of day-

386   to-day PM2.5 variations in spring and autumn lies between those of summer and winter. A total of

387   365 decay processes were recognized from January 2014 to March 2020 based on the regional

388   variation in the day-to-day PM2.5 concentration difference. 97 of the 365 decay phases were related

389   to the effective wet deposition, and most of them occurred in summer. For the dry-day decay

390   processes, 105, 21, 56 and 109 cases occurred in spring, summer, autumn and winter, respectively.

391   The intervals between two continuous decay processes are 5.53, 7.45, 5.86 and 5.36 days from

392   spring to winter, respectively.

393   T-mode PCA circulation classification has been applied to the dry-day decay process of the specific

394   season. Two dominant circulation patterns are identified in summer; three circulation types for the

395   other three seasons, in addition, it shows almost the same circulation patterns in the three seasons.

396   The results show that, in all the circulation patterns, a higher than normal surface wind speed, a

397   negative relative humidity anomaly and positive divergence in the PM2.5 horizontal flux benefit the

398   decay processes. However, there are some distinctive features among the different CTs. In spring,

399   autumn and winter, the dominant CTs are controlled by northeasterly, northwesterly and westerly

400   surface wind anomalies, respectively. With the prevailing northeasterly flow in CT1, clean and dry

401   air masses are transported to the BTH region, resulting in the most significant positive divergence

402   of air pollutants. Moreover, in the vertical direction, air pollutants carried by ascending motion are

403   quickly mixed into the free atmosphere with the aid of strong vertical wind shear at the top of the

404   boundary layer. The combined effects of horizontal and vertical diffusion lead to a reduction in

405   PM2.5 concentrations by over 36%. The most frequent pattern for the decay phase in autumn and

406   winter is CT2, in which the BTH region is located in the eastern portion of an anticyclone system

407   with strong northwesterly wind. CT2 has the highest surface wind speed, lowest relative humidity

408   and highest BLH among the three CTs, all of which are favorable for the quick decay of pollution

409   episodes. The PM2.5 concentrations in the 28 pollution channel cities decrease by more than 40%

410   after CT2 occurs. In CT3, the BTH region borders the cyclone system to the northeast, which leads

411   to a prevailing westerly wind anomaly. In addition to the effective zonal divergence in the PM2.5

412   flux, strong horizontal wind shear in the near-surface improves the more uniform vertical

413   distribution of air pollutants in the boundary layer. After the passage of CT3, 26~29% of local air

414   pollutants are typically removed. The two dry-day circulation patterns in summer are similar to the

415   synoptic patterns of CT1 and CT3 in the other three seasons. A dry air mass with a positive BLH

416   anomaly and the effective horizontal outflow of air pollutants are the main reasons for the abrupt

417   decay phases in summer. The average PM2.5 concentrations on decay process days show a

418   significant decreasing trend from 2014 to 2020, which indicates the success of emission mitigation

419   efforts. Emission reductions have led to a 4.3~5.7 $\mu g/(m^3 \cdot yr)$ decrease in PM2.5 concentrations in

420   the 28 pollution channel cities.

421

422   **Code/Data availability:** Daily PM$_{2.5}$ concentration observations at the 28 channel cities were

423   obtained from the website of Ministry of Ecology and Environment of the People's Republic of

424   China (http://106.37.208.233:20035). Daily four times ECMWF ERA5 dataset during 2014 to 2020

425   are   downloaded   from   https://www.ecmwf.int/en/forecasts/datasets/reanalysis-datasets/era5.

426   Atmospheric circulation classification was conducted using European Cooperation in Science &

427   Technology (COST) plan 733 (cost733class software), which can be downloaded at

428   http://cost733.met.no.

429

430   **Author contributions:** XW and RZ designed research. XW, YT and WY performed the analyses

431   and wrote the paper. All authors contributed to the final version of the paper.

432

433   **Competing interests:** The authors declare that they have no conflict of interest.

434

438

439   **Figure captions:**

440

441   Figure 1. Annual mean PM2.5 concentrations in the 28 pollution channel cities of Beijing from 2014

442   to 2019 (units: μg/m^3).

443

444   Figure 2. Time series of daily mean PM2.5 concentrations in the 28 pollution channel cities from

445   January to March 2019 (units: μg/m^3).

446

447   Figure 3. Probability distribution of the relative day-to-day difference of PM2.5 concentrations. The

448   relative difference is based on the PM2.5 concentration on the previous day. The distributions in

449   spring and autumn are combined in the upper panel, and cases in winter and summer are shown at

450   the bottom.

451

452   Figure 4. Monthly cumulative occurrence of the decay processes of pollution episodes. The orange

453   curve indicates the decay process occurrences on dry days. In total, 365 decay processes are

454   identified from January 2014 to March 2020, and 97 of them are associated with precipitation levels

455   greater than 10 mm/day.

456

457   Figure 5. Distribution of the geopotential height (shaded, units: m^2/s^2) and wind fields at 925 hPa

458   for each circulation type. The number over each subplot indicates the occurrence frequency of the

459   specific circulation type. The solid blue box is the location of the domain region covering the 28

460   pollution channel cities.

461

462   Figure 6. Distribution of the geopotential height anomalies (shaded, unit: m^2/s^2) and wind field

463   anomalies at 925 hPa for each circulation type.

464

465   Figure 7. Zonal averaged profile of the distribution of vertical wind shear anomalies in the domain

466   region (shaded, units: m/(s.100 m)) and the vertical and zonal circulation anomalies. The green line

467   indicates the average location of the top of the boundary layer. Zonal wind shear, circulation and

468   boundary layer height are the average values between 34-40° N. The two dashed lines are the eastern

469   and western boundaries of the domain (112 to118° E). The grey region indicates the average altitude

470   between 34-40° N.

471

472   Figure 8. Boxplot of surface wind speed, boundary layer height (BLH), sea level pressure (slp) and

473   relative humidity (RH) for each circulation type. The dashed line indicates the seasonal mean of the

474   specific variables.

475

476   Figure 9. Boxplot of the divergence of PM2.5 flux over the region of 34-40° N and 112-118° E. The

477   daily divergence is calculated based on the Eq.(1). Zonal and meridional components are the first

478   and second terms of the formula.

479

480   Figure 10. Distribution of the daily mean PM2.5 concentrations before and after the occurrence of

481   decay processes of pollution episodes in the 28 pollution channel cities. The hollow box indicates

482   the concentration on the decay phase day, and the solid box is the value on the previous day. The

483   relative differences in the PM2.5 concentrations after the occurrence of decay process are

484   summarized in Table 2. The number at the top of each box indicates the sample size used for the

485   boxplot. The number in the first line is the sample size of the "before" case; and the second line is

486   for the "after" case.

487

488   Figure 11. The day of the maximum PM2.5 concentration during each pollution episode varies with

489   the duration of the developing phase.

490

491   Figure 12. The density plot of the maximum PM2.5 concentration according to the duration of the

492   developing phase of pollution episodes. Daily PM2.5 concentrations are normalized by their

493   monthly mean value to exclude the effects of seasonal and interannual variations in air quality. A

494   warmer color indicates a higher density of scatter. Pentagrams mark the average maximum PM2.5

495   concentration for the specific duration period.

496

497   Figure 13. Variations in the average PM2.5 concentration on all the decay phase days from 2014 to

498   2020. The black hollow circles indicate the mean PM2.5 concentration in each year. The black line

499   is the fitting line based on the mean value. The number in the subplot is the linear trend of the fitting

500   line.

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716

717 Table 1. Frequency of the relative day-to-day PM2.5 difference within the specific range.

Relative Difference (%)	<-80	-80~-60	-60~-40	-40~-20	-20~0	0~40	40~80	80~120	>120
MAM	0.4	4.4	9.0	13.5	17.2	31.6	14.3	5.4	3.8
JJA	0.2	2.2	7.6	15.6	20.7	34.9	12.0	4.3	2.3
SON	1.3	5.2	9.1	12.2	14.8	29.4	15.2	6.7	5.7
DJF	1.9	6.7	9.7	12.5	13.1	25.2	15.3	7.9	7.2

718

719

720 Table 2. The average relative difference of PM2.5 concentrations before and after the occurrence of

721 decay processes (i.e., $(PM_t-PM_{t-1})/PM_{t-1}*100$, where PM_t is the daily mean PM2.5 concentration on

722 the decay phase day).

%	CT1	CT2	CT3	Wet deposition
MAM	-37.2	-44.8	-28.2	-40
JJA	-34.5	-20.4	//	-26.2
SON	-40.1	-42.9	-26.9	-35.8
DJF	-36.9	-41	-29.3	-43.9

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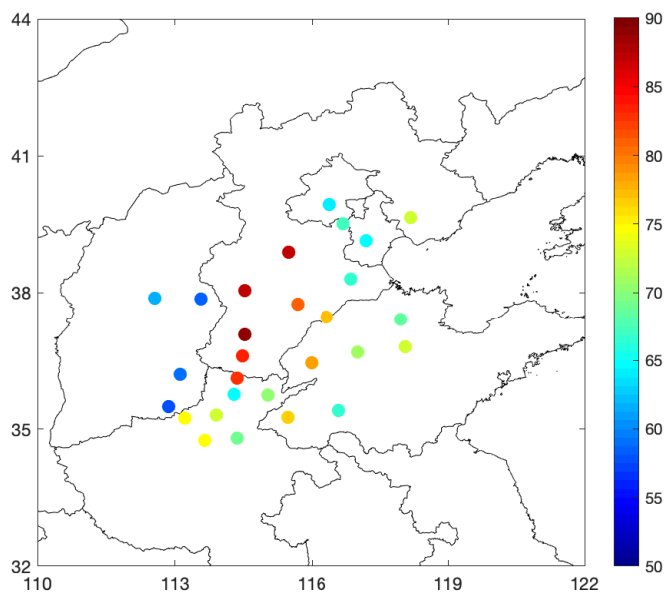
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727 Figure 1. Annual mean PM2.5 concentrations in the 28 pollution channel cities of Beijing from 2014

728 to 2019 (units: μg/m^3).

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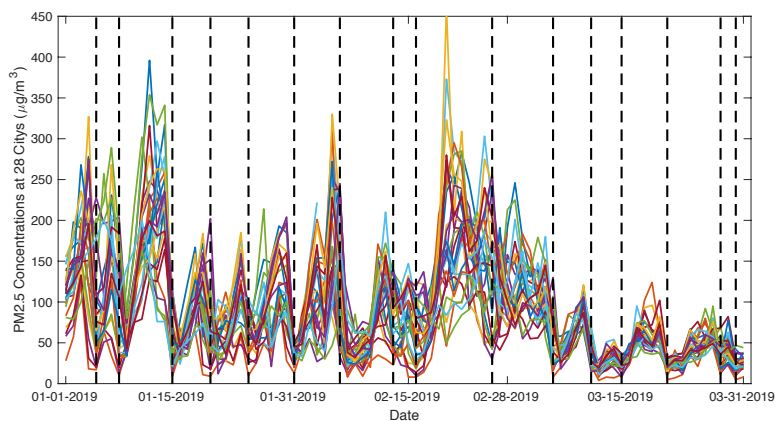
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731 Figure 2. Time series of daily mean PM2.5 concentrations in the 28 pollution channel cities from

732 January to March 2019 (units: μg/m^3).
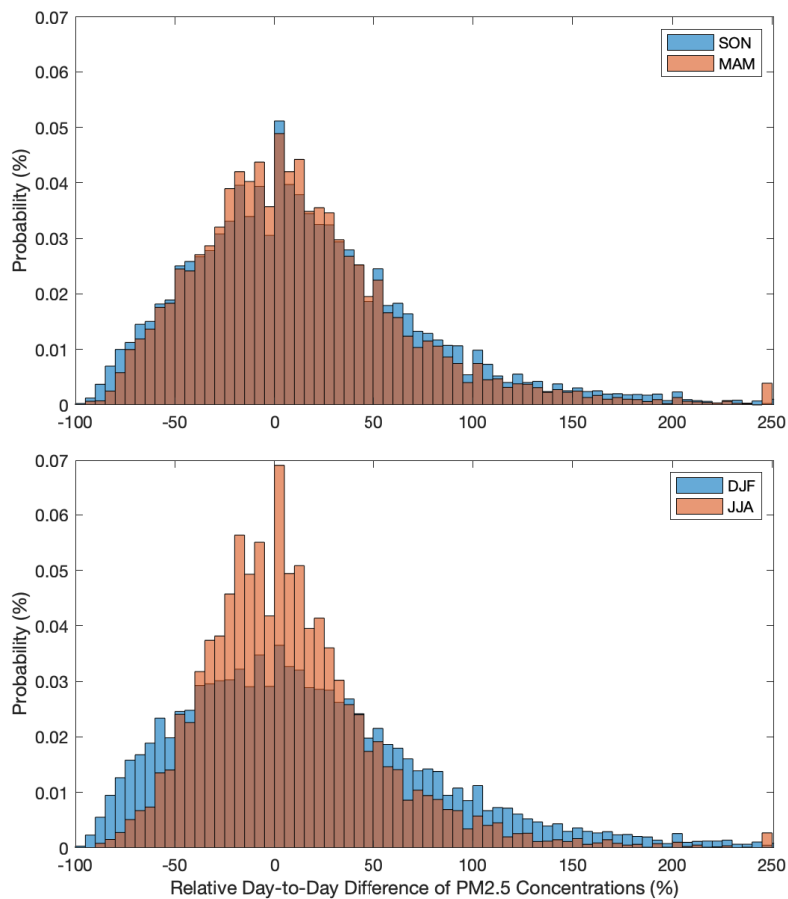
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Figure 3. Probability distribution of the relative day-to-day difference of PM2.5 concentrations. The relative difference is based on the PM2.5 concentration on the previous day. The distributions in spring and autumn are combined in the upper panel, and cases in winter and summer are shown at the bottom.
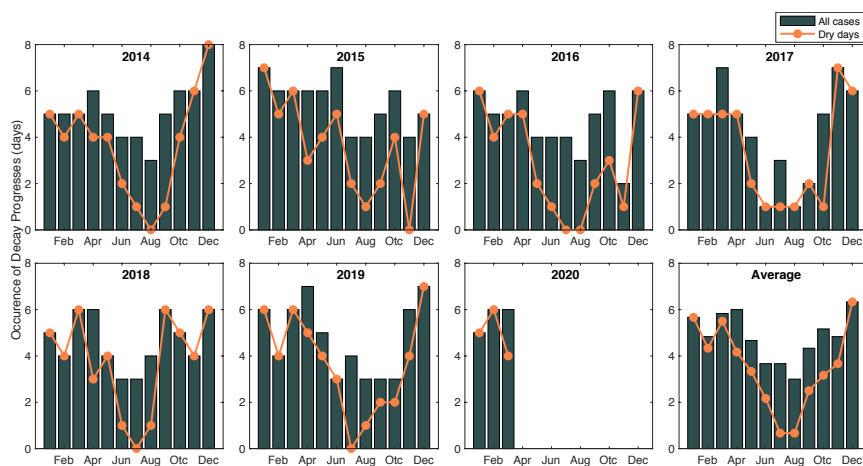
739

740   Figure 4. Monthly cumulative occurrence of the decay processes of pollution episodes. The orange

741   curve indicates the decay process occurrences on dry days. In total, 365 decay processes are

742   identified from January 2014 to March 2020, and 97 of them are associated with precipitation levels

743   greater than 10 mm/day.

744

Figure 5. Distribution of the geopotential height (shaded, units: m^2/s^2) and wind fields at 925 hPa for each circulation type. The number over each subplot indicates the occurrence frequency of the specific circulation type. The solid blue box is the location of the domain region covering the 28 pollution channel cities.

Figure 6. Distribution of the geopotential height anomalies (shaded, unit: m²/s²) and wind field anomalies at 925 hPa for each circulation type.

Figure 7. Zonal averaged profile of the distribution of vertical wind shear anomalies in the domain region (shaded, units: m/(s.100 m)) and the vertical and zonal circulation anomalies. The green line indicates the average location of the top of the boundary layer. Zonal wind shear, circulation and boundary layer height are the average values between 34-40° N. The two dashed lines are the eastern and western boundaries of the domain (112 to118° E). The grey region indicates the average altitude between 34-40° N.

760

Figure 8. Boxplot of surface wind speed, boundary layer height (BLH), sea level pressure (slp) and

relative humidity (RH) for each circulation type. The dashed line indicates the seasonal mean of the

specific variables.

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Figure 9. Boxplot of the divergence of PM2.5 flux over the region of 34-40° N and 112-118° E. The

daily divergence is calculated based on the Eq. (1). Zonal and meridional components are the first
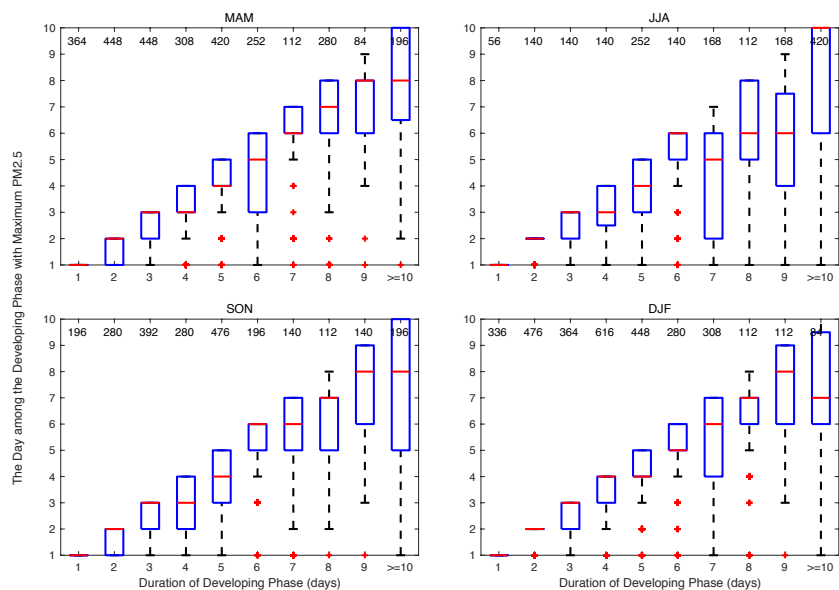
and second terms of the formula.

768

Figure 10. Distribution of the daily mean PM2.5 concentrations before and after the occurrence of decay processes of pollution episodes in the 28 pollution channel cities. The hollow box indicates the concentration on the decay phase day, and the solid box is the value on the previous day. The relative differences in the PM2.5 concentrations after the occurrence of decay process are summarized in Table 2. The number at the top of each box indicates the sample size used for the boxplot. The number in the first line is the sample size of the "before" case; and the second line is for the "after" case.

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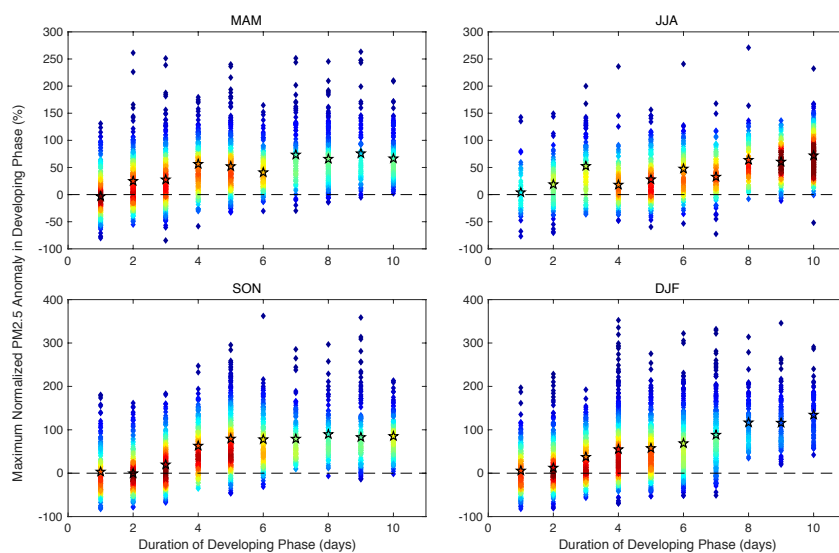777

778 Figure 11. The day of the maximum PM2.5 concentration during each pollution episode varies with
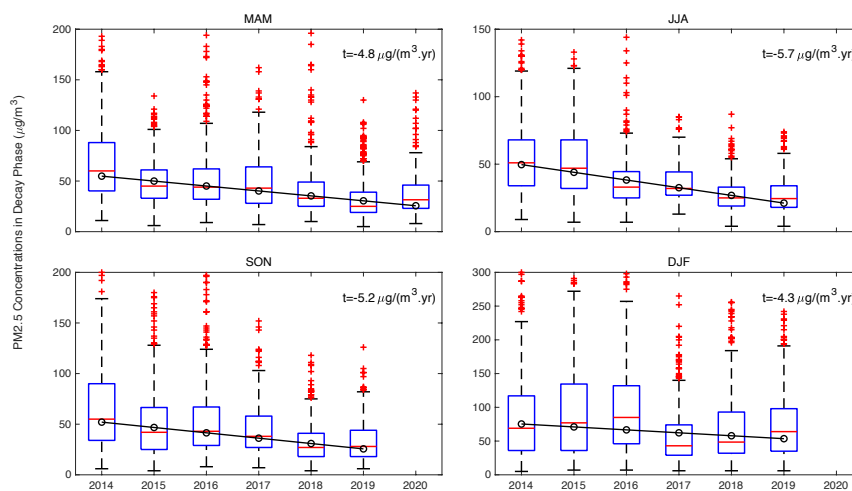
779 the duration of the developing phase.

780

Figure 12. The density plot of the maximum PM2.5 concentration according to the duration of the developing phase of pollution episodes. Daily PM2.5 concentrations are normalized by their monthly mean value to exclude the effects of seasonal and interannual variations in air quality. A warmer color indicates a higher density of scatter. Pentagrams mark the average maximum PM2.5 concentration for the specific duration period.

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Figure 13. Variations in the average PM2.5 concentration on all the decay phase days from 2014 to
2020. The black hollow circles indicate the mean PM2.5 concentration in each year. The black line
is the fitting line based on the mean value. The number in the subplot is the linear trend of the fitting
line.

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