



Synoptic drivers of co-occurring summertime ozone and PM2.5 pollution in eastern China

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Abstract

- In recent years, surface ozone (O₃) pollution during summertime (June-August) over eastern China has become more serious, and it is even the case that surface O₃ and PM_{2.5} (particulate matter with aerodynamic diameter ≤ 2.5 µm in the air) pollution can co-occur. However, the synoptic circulation pattern related to this compound pollution remains unclear. In this study, the T-mode principal component analysis method is used to objectively classify four synoptic weather patterns (SWPs) that occur over eastern China, based on the geopotential heights at 500 hPa during summertime from 2015 to 2018. Four SWPs of eastern China are closely related to the western Pacific subtropical high (WPSH), exhibiting, significant intraseasonal and interannual variations.
 - Note that remarkable spatial and temporal disparities of surface O₃ and PM_{2.5} pollution are given under these four different SWPs according to the ground-level air quality and meteorological
- 30 observations. In areas controlled by the WPSH or the prevailing westerlies, O₃ pollution is mainly caused by photochemical reactions of nitrogen oxides and volatile organic compounds under weather conditions of high temperature, moderate humidity and slight precipitation. In particular, the warm moist flow brought by the WPSH can promote hygroscopic growth of fine particulate matter in some local areas, resulting in the increase of PM_{2.5} concentrations, which may form co-
- 35 occurring surface O₃ and PM_{2.5} pollution. In addition, the low boundary layer height and frequency of light-wind days are closely related to the transmission and diffusion of pollutants under the different SWPs, modulating the levels of O₃-PM_{2.5} compound pollution. Overall, our findings demonstrate the different roles played by synoptic weather patterns in driving regional surface O₃-PM_{2.5} compound pollution, in addition to the large quantities of emissions, and may also provide
- 40 insights into the regional co-occurring high PM_{2.5} and high O₃ level via the effects of certain meteorological factors.

Keywords: Synoptic weather pattern, ozone, PM_{2.5}, compound pollution, western Pacific subtropical high (WPSH)

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1. Introduction

In recent years, China has been experiencing serious air pollution problems due to the





enormous emissions of polluting gases [e.g., sulfur dioxide, nitrogen dioxide (NO₂), etc.] and aerosol particulates [e.g., particulate matter with aerodynamic diameter ≤ 2.5 (10) µm in the air
PM_{2.5} (PM₁₀), etc.] associated with its rapid economic development, industrialization and urbanization, together with certain unfavorable meteorological conditions (Wang & Chen, 2016; Zhang et al., 2014; Y. Zhang et al., 2016). Particularly, atmospheric compound pollution has become serious (Li et al., 2019; Saikawa et al., 2017; C. Zhang et al., 2019), especially for the economically developed and densely populated eastern urban agglomerations of China, such as the Beijing–

55 Tianjin–Hebei (BTH), Yangtze River Delta (YRD) and Pearl River Delta (PRD) regions (Cai et al., 2017; Du et al., 2019; Ji et al., 2018; Li et al., 2020), exerting a severe threat in terms of public health, economy and society (Chen et al., 2019; Cohen et al., 2017; Day et al., 2017; Yim et al., 2019).

In general, PM2.5 pollution is featured with obvious diurnal and seasonal changes. Due to the

- 60 influence of atmospheric diffusion conditions such as precipitation and wind speed (WS), it tends to be enhanced in the morning and evening, lower at noon, and higher in winter and lower in summer (Amil et al., 2016; H. Liu et al., 2019; Ye et al., 2018; Zhang & Cao, 2015). The PM_{2.5} level of China showed a steady increase from 2004 to 2007, and has since stabilized (Ma et al., 2016); however, there are still frequent PM_{2.5} pollution events in autumn and winter (Song et al., 2017; Yang et al.,
- 65 2018; Ye et al., 2018; Zhang et al., 2014). In the past few years, the PM_{2.5} concentration in China has decreased significantly as a result of measures introduced across the country that have reduced multi-pollutant emissions, adjusted energy structure, and increased supply of clean energy (Gui et al., 2019; Yang et al., 2020; Q. Zhang et al., 2019; Zhang et al., 2020). In contrast, summer O₃ pollution has gradually been prominent, replacing PM_{2.5} as the primary pollutant in the air (Li et al.,
- 2019). As a secondary pollutant, ozone in the troposphere is mainly formed by photochemical reactions between NO_x, carbon monoxide (CO) and VOCs in the exposure of sunlight (Sillman, 2002). The prominent problem of O₃ pollution has attracted the attention of experts and scholars in recent years. For instance, Sun et al. (2016) showed that the observed summertime O₃ at Mt. Tai has increased significantly by 1.7 ppbv yr⁻¹ for June and 2.1 ppbv yr⁻¹ for July–August during the period of 2003 to 2015. An increase of the maximum daily 8-h average concentration of O₃ (MDA8 O₃) at

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frequency of PM_{2.5} pollution.

Many studies have indicated that PM_{2.5} and O₃ pollution are strongly correlated with local meteorological factors such as temperature, relative humidity (RH) and WS (Huang et al., 2016;

- 80 Miao et al., 2015; Shu et al., 2019; Tai et al., 2010). Miao et al. (2015) suggested that strong northwesterly synoptic winds, low BLH (boundary layer height), high RH and stable atmosphere are more prone to aerosol pollution in the BTH region during wintertime. Zhang et al. (2017) found that the majority of O₃ extremes occurred with daily maximum temperature (Tmax) between 300 K and 320 K, minimum RH (RHmin) less than 40%, and minimum WS less than 3 m s⁻¹, through the
- 85 analysis of extreme O₃ and PM_{2.5} events from historical data (30 years for O₃ and 10 years for PM_{2.5}) in the US. Furthermore, the number of annual extreme PM_{2.5} days was highly positively correlated with the extreme RHmin/Tmax days, and the correlation coefficient between PM_{2.5} and RHmin (Tmax) was highest in urban and suburban (rural) regions. Shi et al. (2020) studied the spatial distribution of O₃-8h (O₃ 8-hour moving average) and PM_{2.5}, and their sensitivity of meteorological
- 90 parameters; pronounced positive (negative) correlation between temperature (BLH and absolute humidity) and O₃-8h was found, but the relation between WS and O₃-8h was spatially different; for PM_{2.5}, it was negatively (positively) correlated with temperature, WS and BLH (absolute humidity). Recently, Han et al. (2020) revealed that meteorological factors can explain ~46% of the daily variability in summertime surface O₃, while synoptic factors contribute to ~37% of the overall
- 95 meteorological effects on the daily variability of surface O₃ in eastern China. The abovementioned indicates that the variation of meteorological factors, which are mainly driven by the evolution of different weather circulation situations, play a non-negligible role in air pollution. Therefore, classification of air pollution according to the meteorological circulation has become particularly important, not least because of its worth when applied to air quality monitoring, forecasting and

100 evaluation (Liu et al., 2019; Ning et al., 2019; Yang et al., 2018; Zheng et al., 2015).

In recent years, it has become possible to objectively classify atmospheric circulation conditions using weather data such as GH, sea level pressure, WS and temperature, so that the weather mechanism of extreme weather can be better understood and analyzed. Compared with subjective weather classification, the objective approach has been widely used in air pollution research (Beck & Philipp, 2010; Miao et al., 2017, 2019; Ning et al., 2018). Miao et al. (2019),





based on the daily 900 hPa GH fields during winter in Beijing, identified seven synoptic patterns using an objective approach, and found that the weak northwesterly prevailing winds and strong elevated thermal inversion layer, along with the local emissions of aerosols play a decisive role in the formation of heavy pollution in Beijing; noted also that the southerly prevailing winds can

- 110 transport the pollutants emitted from southern cities to Beijing. Zheng et al. (2015) studied the relationship between regional pollution and the patterns of large-scale atmospheric circulation over eastern China in October from 2001 to 2010 and identified six pollution types and three clean types. Specifically, weather patterns such as a uniform surface pressure field in eastern China or a steady straight westerly in the middle troposphere, particularly when at the rear of an anticyclone at 850
- 115 hPa, were found to be typically responsible for heavy pollution events. Many studies have suggested that PM_{2.5} and O₃ pollution are mainly related to the East Asian summer monsoon (EASM) and western Pacific subtropical high (WPSH) (Li et al., 2018a; Xie et al., 2017; Yin et al., 2019; Zhao et al., 2010). The anomalous high-pressure system at 500 hPa, associated with downward dry, hot air and intense solar radiation can enhance the photochemical reactions to elevate the production of
- 120 tropospheric O₃ (Gong and Liao, 2019; Yin et al., 2019). Furthermore, Zhao & Wang (2017) and Yin et al. (2019) noted that the positive GH anomalies at high latitudes tend to significantly weaken the cold-air advection from the north and result in local high temperatures near the surface in northern China, while the WPSH can transport sufficient water vapor to the YRD region and lead to a decrease in surface O₃. In addition, different subregions can exhibit various distributions of
- 125 pollutant, even with identical emission scenarios (Li et al., 2019; Saikawa et al., 2017; C. Zhang et al., 2019). Also, it is still unclear how the distribution of pollution responds locally to large-scale atmospheric circulation patterns. Considering the reduced surface solar radiation due to PM_{2.5} pollution (Haywood & Boucher, 2000; He & Wang, 2020; Huang et al., 2018; Yang et al., 2020) and thus suppressed the photochemical production of O₃ (Li et al., 2017), the compound O₃-PM_{2.5}
- 130 pollution-related meteorological conditions, should be complex and likely to be associated with certain weather types. Overall, the mechanism by which the synoptic weather pattern (SWP) modulates the characteristics of O₃-PM_{2.5} compound pollution has yet to be comprehensively described.

In this study, the SWPs corresponding to the co-occurrence O₃-PM_{2.5} pollution during





135 summertime are analyzed, focusing on the eastern China (108°–135°E, 17°–53°N). Then, the causes of O₃-PM_{2.5} compound pollution, as well as O₃-only pollution, from the perspective of the objective classification of atmospheric circulation patterns, are revealed. The findings are expected to provide a scientific reference for the monitoring, forecasting and evaluation of summertime air pollution in eastern China.

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2. Data and methods

The air quality data, including $PM_{2.5}$, NO_2 , O_3 , and O_3 -8h, are from the national 24-h continuous air quality observation published by the China Environmental Monitoring Station (<u>http://www.cnemc.cn/</u>). The hourly observation data of a total from 949 stations (108° - $135^{\circ}E$, 17° -

- 145 53°N) in eastern China during summertime of 2015–2018, which include the more prominent pollution areas in the eastern urban agglomeration, such as the BTH, YRD, PRD, Guanzhong Plain (GZP), Northeast Megalopolis (NEM) regions (the specific locations of stations and urban agglomerations are shown in Fig. 1a). Surface meteorological data, such as Tmax, precipitation, WS and RH from 517 meteorological observation stations and radiosonde data from 63 stations in
- 150 eastern China, were obtained from the China National Meteorological Information Center of the China Meteorological Administration (<u>http://data.cma.cn/site/index.html</u>). The BLH was calculated according to the method given by Guo et al. (2016, 2019), and the FLWD [frequency of light wind (< 2 m s⁻¹) days], precipitation frequency (PF), and MDA8 O₃ were also counted.

Additionally, for synoptic analysis of particulate matter and O₃ pollution in summer, we use 155 the GH field at 500 hPa and wind field at 850 hPa from the NCEP/NCAR (National Centers for Environmental Prediction/National Center for Atmospheric Research) daily reanalysis dataset on a 2.5° × 2.5° latitude/longitude grid during the study period.

The T-mode principal component analysis (T-PCA) is an objective mathematical computerbased method that can be used to classify the synoptic circulation patterns of regional gridded data in the troposphere at the lower level. Indeed, it is commonly regarded as the most promising weather pattern classification method at present (Huth et al., 2008). Moreover, this approach has been widely used in the studies of aerosols and O₃ pollution-related atmospheric circulation in China (Miao et al., 2017, 2019; Ning et al., 2018, 2019). The T-PCA analysis module of the COST733 software





(http://cost733.met.no/) developed by the European Scientific and Technical Research Cooperation, was used to classify the synoptic circulation pattern based on the 500 hPa GH field. More detailed information about the T-PCA method can be found in Miao et al. (2017). To assess the performance of synoptic classification and determine the number of classes, the explained cluster variance (ECV) is selected in this study (Hoffmann & SchlüNzen, 2013; Ning et al., 2019; Philipp et al., 2014). The detailed information about the ECV is provided in the supplementary document.

- 170 Based on the GB3095-2012 environmental air quality standard issued by the Ministry of Ecology and Environment of China, O₃ (PM_{2.5}) pollution occurs when the MDA8 O₃ exceeds 160 (75) μg m⁻³. To investigate the temporal variations of air pollution in various regions in summer, the pollution levels of O₃ and PM_{2.5} in each key area were verified according to their concentration limits.
- 175 Finally, in order to make it clear in the analysis of different weather types of O₃ and PM_{2.5} concentration change, we calculated the average distribution of O₃ and PM_{2.5}, as well as the meteorological conditions for each type, and further calculated the anomalous distribution of these variables, i.e., the average of O₃ and PM_{2.5} and the average of the meteorological conditions under the respective patterns minus the average during summertime of 2015–2018, were given as well.
- 180 The statistical significance was tested with a 0.05 confidence level via analysis of variance, which enabled us to distinguish the significant differences of spatial distribution characteristics between O₃ and PM_{2.5} pollution under four SWPs.

3. Results

185 3.1 Spatial and temporal distribution of O₃ and PM_{2.5} during summer 2015–2018

Figure 1 shows the summer averaged MDA8 O₃ and PM_{2.5} concentrations at 949 stations in the eastern region of China for 2015–2018. Among these stations, the MDA8 O₃ concentration at most stations (662/949) exceeds 100 µg m⁻³, of which 45 sites exceed 160 µg m⁻³. The highest O₃ pollution is found in Zibo, Shandong, with a value of 181.5 µg m⁻³. The averaged PM_{2.5} at most sites (680/949) is below 35 µg m⁻³, while reaches 62.6 µg m⁻³ in Handan, Hebei Province. On the whole, the MDA8 O₃ and PM_{2.5} in the BTH region and its surrounding areas is significantly higher than in other regions; and besides, the level of O₃ in some urban clusters, such as the PRD, YRD,





GZP and NEM regions, is particularly higher than that of the surroundings, thus, we focus on analyzing these key areas later.

- The temporal variations of O₃ pollution levels in key areas are displayed in Fig. 2, revealing an obvious increase in O₃ pollution levels and duration in the five priority areas for the period 2015–2018. The O₃ pollution in June is more severe than that in July and August, which is consistent with the fact that the peak concentration of MDA8 O₃ in northern China is in June (Gong and Liao, 2019). In spatial terms, O₃ pollution is particularly prominent in the BTH and GZP regions, with the number
- 200 of pollution days reaching 148 and 109, of which even 23 and 17 are moderate pollution, respectively. Figure 3 illustrates the PM_{2.5} pollution-level variations and, although it is not difficult to see that PM_{2.5} pollution weakens year by year, the number of days of PM_{2.5} pollution in the BTH region reaches 192 (25 days for moderate pollution), which is higher than the number of days of O₃ pollution. The reduced visibility of haze days weakens the solar radiation reaching the ground and
- 205 inhibits photochemical reactions from generating O₃ (Li et al., 2019; Z. Zhang et al., 2015), as a result, the concentration of O₃ continues to increase with the mitigation of PM_{2.5} pollution. It is worth noting that O₃ and PM_{2.5} co-occurred in both the BTH (120 days) and GZP (60 days) regions. Overall, the O₃ and PM_{2.5} concentration in eastern China exhibits distinct intraseasonal and interannual variations, indicating that, aside from the changes in emission sources (because it is
- 210 considered that inter-seasonal and short-term changes in emission sources are not significant), it may also be regulated by meteorological conditions, which is further analyzed below.

3.2 Objective classification of large-scale synoptic circulation patterns in summer

To analyze the effect of meteorological conditions on the changes of O₃ and PM_{2.5} 215 concentration, it is necessary to statistically analyze the large-scale weather circulation situation in summer. Existing studies have shown that the WPSH (500 hPa GH field with obvious anticyclonic characteristics, and downward flow around the center) in summer prominently regulates the weather and climate of East Asia (Lu, 2002), owing to its various location, shape and intensity (Ding, 1994). A low-level southerly monsoon formed at the periphery of the WPSH can transport warm and humid 220 air from the ocean to East Asia, which might also be responsible for the asymmetric spatial

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but an increase over northern China (Zhao & Wang, 2017).

Therefore, we used the T-PCA method to objectively classify the weather circulation of the 500-hPa GH field in the summers of 2015–2018, and finally obtained four SWPs related to the

- 225 movement and development of the WPSH. The westward extension and southward motion of the WPSH in Type 1, as shown in Fig. 4a, transports water vapor into the YRD region, and the prevailing southwesterly in the YRD region and westward flow from the north form a cyclonic convergence area, with high temperature and high humidity during the Meiyu season. For Type 2, it is noticed that the westerly trough deepens accompanying the northward (or southward) advance (retreat) of
- 230 WPSH, and the GH over northern China at 500 hPa is higher compared with Type 1 (Fig. 4b). The southerly wind blowing from the ocean to the continent lies in front of the bottom of the high pressure, affecting the southeastern region, while northern China is mainly controlled by the westerly trough, and the rain belt moves northwards to the east of the YRD region. Under Type 3, the WPSH shifts further north with a westward extension, and disintegrates a closed high-pressure
- 235 monomer along the eastern coast of China, while the main body of the WPSH remains on the ocean (Fig. 4c). In this case, the YRD region is completely controlled by the monomer of the WPSH, which implies that the rainy season in the YRD region ends in midsummer and the weather becomes hot and dry. At the same time, the rain belt moves gradually northwards to the BTH and NEM regions. According to Fig. 4d, the monomer of WPSH under Type 4 continues to extend westwards
- 240 and shift northwards, controlling northern China for a long time, and with persistent high temperatures and a heat wave occurring in most parts of the eastern China.

Fig. 5 presents the daily and annual variations of the SWPs in the summers of 2015–2018. The advance of the WPSH in eastern China occurs in June and July, while gradual withdrawal occurs mainly in August, so Type 1 mainly appears in June, while Type 2, Type 3 and Type 4 occurs mainly in July and August. Consequently, Type 1, Type 2, Type 3 and Type 4 appear for 167, 117, 52 and 32 days, respectively, during the study period. Since the movement of the WPSH is often affected

by the activities of the surrounding weather system (such as typhoons, the Tibetan high, etc.) (Ge et al., 2019; Liu & You, 2020; Shu et al., 2016), there may be a short southward retreat during the WPSH's advancement (e.g., around 10 August 2018) and a short northward advance during its process of retreat (e.g., 21 and 29 August 2016). In general, it can be seen that the WPSH also shows





evident intra-seasonal and interannual changes, which will inevitably regulate the weather, climate and environment changes in eastern China.

3.3 O₃ and PM_{2.5} pollution characteristics under four SWPs

255 3.3.1 Spatial characteristics

We calculated the averaged and anomalous spatial distributions of the MDA8 of O₃ and PM_{2.5} under the four SWPs. The averaged MDA8 O₃ under the four SWPs can be seen in Fig. S2. The O₃ concentration is relatively high in the area north of the Yangtze River under Type 1, and the high values of the MDA8 O₃ are mainly concentrated in the North China Plain (NCP) region, with a total

- of 94 stations surpassing 160 μ g m⁻³. Type 2 O₃ pollution is slightly weaker than that for Type 1, and the MDA8 O₃ at the 72 sites exceeds 160 μ g m⁻³. The O₃ high-value areas lie mainly in the NCP, GZP and YRD regions under Type 4, and there are 37 stations larger than 160 μ g m⁻³. Of the four SWPs, the lowest overall MDA8 O₃ occurs under Type 3, with only one site exceeding 160 μ g m⁻³. It is also found that the regions experiencing significant positive deviations of the MDA8 O₃
- 265 from the summer mean are as follows: the BTH, YRD and NEM regions under Type 1, the BTH and GZP regions under Type 2, the middle of the YRD and PRD regions under Type 3, and the YRD, GZP and PRD regions under Type 4 (Fig. 6).

Analogously, Fig. 7 shows the anomaly and significance of difference of PM_{2.5} under the four weather types, presented as positive anomalies in the south of the BTH and YRD regions under Type

270 1, in the BTH, GZP and PRD regions under Type 2, and in the GZP and PRD regions under Type 4. Due to the obvious seasonal variations of PM_{2.5} concentration (higher in winter and lower in summer) (H. Liu et al., 2019; Miao et al., 2015), no site exceeds 75 µg m⁻³ for the averaged PM_{2.5} concentration. Even so, the level of PM_{2.5} in the BTH region is still significantly higher under the four types than that for the other urban agglomerations (Fig. S3).

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3.3.2 Pollution pattern differences in key areas

Air pollution in eastern China is principally found in dense urban areas such as the BTH and YRD regions (Gui et al., 2019; Han et al., 2019), so we took the BTH, PRD, YRD, GZP and NEM regions in the eastern region as key areas, counted the average daily changes of O₃ and PM_{2.5} in





- 280 each key region under different weather patterns (Fig. 8), and calculated the over-limit ratio in key regions via the stations × days statistics (see Table 1). The diurnal variation of O₃ is more obvious, peaking at about 15:00 (Beijing time), while contrasting diurnal variations of PM_{2.5} are given for different regions. According to Figure 8 and Table 1, the following characteristics can be identified for different urban clusters: (1) in the BTH region, the O₃ pollution of Type 1 and Type 2 is relatively
- 285 serious, their over-standard rates reach 47.1% and 54.2%, and the PM_{2.5} pollution rates of Type 2 and Type 1 reach 18.8% and 16.3%, respectively; (2) in the PRD region, the over-standard rates variation of O₃ and PM_{2.5} is equable; (3) in the YRD region, the O₃ pollution over-limit ratio presents as Type 1 > Type 4 > Type 2 > Type 3, PM_{2.5} pollution largely appears in Type 1, and both O₃ and PM_{2.5} in Type 1 are higher than those in the other types; (4) in the GZP region, the O₃ pollution
- 290 frequency is higher in Type 2 and Type 4, and PM_{2.5} pollution occurs more frequently in Type 2; and (5) in the NEM region, O₃ pollution is always found in Type 1, Type 2 and Type 4, but the overstandard rate is no more than 15% and PM_{2.5} pollution in Type 1 is more than in Type 2.

In summary, Type 1 is prone to the formation of compound pollution of O_3 -PM_{2.5} (that is, when the ground MDA8 O_3 concentration exceeds 160 µg m⁻³, the PM_{2.5} concentration also exceeds 75

295 μg m⁻³) in the southern BTH and northern YRD regions, which can be denoted as "South BTH – North YRD O₃-PM_{2.5} compound pollution". Similarly, Type 2 can be denoted as "BTH – GZP O₃-PM_{2.5} compound pollution", Type 3 as "BTH – GZP O₃-only pollution", and Type 4 as "GZP O₃-PM_{2.5} compound pollution with BTH – YRD – PRD O₃-only pollution".

300 4. Discussion

4.1 Analysis of potential meteorological factors

The activities of atmospheric circulation system often lead to changes in meteorological elements, and to a large extent, affect the processes of pollutant formation, transmission and diffusion. Zhang et al. (2017) revealed that the extreme O₃ and PM_{2.5} pollution events in the United 305 States always occur under the conditions of high temperature, low humidity and low WS, while Miao et al. (2015) showed that RH is high when aerosol pollution occurs in the BTH region. However, O₃ pollution in China is more frequent in summer, and the warm and humid flow brought by the EASM makes summer always hot and moist. Zhao et al. (2019) investigated the RH of O₃





pollution in Shijiazhuang between 15 June and 14 July 2016, and found that the O₃ concentration
was higher at moderate humidity (RH average during daytime from 10:00 to 17:00 LT was 40%–50%).

Therefore, to explore the meteorological causes of O_3 and $PM_{2.5}$ pollution, we analyzed the distribution of the average and anomalies for Tmax, RH, PF, BLH and FLWD under the four SWPs (Figs. S4, S5, 9 and 10). Under the influence of the EASM, over 80% of the stations experience

- 315 high temperatures (Tmax > 27°C) in each SWP, although the anomaly of Tmax in Type 1 presents negative. Type 1 is characterized by humid condition in the south and dry condition in the north, with the rain belt mainly in the PRD and YRD regions; Type 2 has dry and wet anomalies meridionally in northern China, and the rain band is located in the middle of the BTH and YRD regions; the RH is large for most sites under Type 3 and Type 4, corresponding to the shifted rain
- 320 belt to the BTH and NEM regions under Type 3, and the heavy precipitation appeared in the western PRD region and the middle of the BTH and YRD regions under Type 4 (Fig. S4).

In terms of their anomalous spatial distribution, the positive anomalies of Tmax are located in the southern region of Type 3 and most of the eastern region of Type 4; and since Type 1 always appears in June, most areas are negative (Figs. 9a–d). For RH, Types 2, 3 and 4 are negative for the

- 325 south and positive for the north, while Type 1 is opposite (Figs. 9e–h). PF is characterized by positive anomalies in the area south of the Yangtze River under Type 1, in the YRD region under Type 2, in the BTH and NEM regions under Type 3, and in the area between the BTH and YRD regions under Type 4 (Figs. 9i–l). As can be seen from Fig. 10, when the BLH has a positive anomaly, on the contrary FLWD has a negative anomaly, which indicates the higher the height of the boundary layer,
- 330 the lower the frequency of light wind days, the more conducive to the diffusion of pollutants, and vice versa. After further inspection of Fig. S4, we found the YRD region in Type 1, the area north of the Yangtze River in Type 2, the BTH and PRD regions in Type 3, and most regions of Type 4 have shallow BLHs and high FLWDs, which is detrimental to the transportation of pollution in these areas, thus corresponding to high levels of pollution under these weather patterns.

335 **4.2 Effects of NO₂ on O₃**

Photochemical production of O_3 mainly involve emissions of VOCs and NO_x from anthropogenic, biogenic and biomass burning sources (Deng et al., 2019; Gvozdić et al., 2011;





Sillman, 2002). Due to the implementation of a number of pollution abatement measures, the abundance of NO₂ has decreased significantly in eastern China in the past few years, leading to the
sensitive precursor of O₃ formation changing from VOCs to a mixture of VOCs and NO_x (Wang et al., 2019b). As a result, both NO_x and VOCs need being cooperatively controlled to mitigate O₃ pollution. Figure 11 shows the diurnal variations of NO₂ and the ratio of O₃ to NO₂ (O₃/NO₂) in five urban clusters, where the larger the O₃/NO₂ ratio, the more O₃ is generated by the photochemical reaction of NO₂. The NO₂ concentration reaches its lowest and O₃/NO₂ reaches a peak at around

- 345 15:00 (Beijing time) in the day, owing to the rapid consumption of NO₂ by the photochemical reaction under high temperature and strong solar radiation in the afternoon. As far as O₃/NO₂ (NO₂) is concerned, it can be seen that the daytime O₃/NO₂ (NO₂) values under Type 4 in the five regions is greater than (less than) under the other types, indicating that O₃ photochemical reactions are stronger under this type than the others. In contrast, the photochemical reaction of the BTH and
- 350 NEM regions under Type 3 and the YRD region under Type 1 is weaker owing to the warm and humid air brought by the WPSH, and the rainy weather is also unconducive to the occurrence of photochemical reactions to generate O₃. For the PRD region, the photochemical processes of the four SWPs are not significantly different. It can be seen that different weather patterns also have an important regulatory effect on the photochemical production of O₃.
- 355 In summary, the different SWPs can modulate the regional variability of summertime O₃ and PM_{2.5} and their causes in summer as follows:

(1) Type 1: The area to the north of the Yangtze River under Type 1 is controlled by the westerly zone in the north of the WPSH at 500 hPa. Under the conditions of high temperature (Tmax > 27°C), moderate humidity (RH ~60%), and low PF, photochemical reactions are largely promoted to cause severe O₃ pollution. Meanwhile, the area from south of the BTH region to north of the YRD region is located in front of the westerly trough, under the influence of the warm and humid air of the WPSH, and so the hygroscopic growth of fine particulates will cause a certain amount of PM_{2.5} pollution, becoming O₃-PM_{2.5} compound pollution (Fig. 12). In Figure S6, we counted the number and probability of occurrence of compound pollution days in each site in summer during 2015-2018, indicating that high occurrence probability (maximum values can approach 46.7%) of compound





pollution occurrence days at all sites occurred under Type 1.

(2) Type 2: The westerly trough strengthens meridionally, and the northern region is still controlled by the westerly zone. Ozone pollution is severe under the meteorological conditions of

- 370 high temperature, moderate humidity, and few precipitations. The PM_{2.5} in the BTH region, which is located in front of the westerly trough, is high since the shallow boundary layer and low wind frequency are unfavorable for pollutant diffusion. Therefore, O₃-PM_{2.5} compound pollution can also be rather frequent (Fig. 12). About 33.0% of compound pollution occurrence days at all sites occurred under Type 2 in summer during 2015-2018.
- 375 (3) Type 3: High temperature, low humidity and few precipitations in the YRD region generates a large amount of O₃, while the positive BLH and negative FLWD anomalies are unfavorable to O₃ accumulation. On the other hand, summer typhoon activities will weaken the WPSH intensity over the YRD region, leading to the eastward retreat and northward shift of the WPSH, high WS in coastal areas that will ease O₃ pollution on the ground (Shu et al., 2016), and
- 380 high PF in the BTH and PRD regions which tend to suppress O₃ production. Accordingly, Type 3 is characterized as light O₃-only pollution in the areas of the BTH, YRD and PRD (Fig. 12).

(4) Type 4: High temperatures, medium-high humidity and few precipitations in the GZP and PRD regions can cause O₃-PM_{2.5} compound pollution, but PM_{2.5} pollution in the PRD region is not heavy, which is possibly in relation to local lower pollutant emissions. Under the control of the

- 385 WPSH, there are strong photochemical reactions at high temperatures and little rainfall in the BTH region, which is also conducive to O₃ generation (Fig. 12). Meanwhile, relative to Type 1, O₃ pollution is lighter in the BTH, due to the differences of RH, BLH and FLWD.
 - 5 Conclusions

In this study, T-PCA, an objective classification method, was applied to classify the 500-hPa 390 weather circulation pattern as four SWPs in the summers of 2015–2018. It was found that these four SWPs are closely related to the development of the WPSH. The spatial and temporal distribution characteristics of O₃ and PM_{2.5} pollution in eastern China under the four SWPs were analyzed to regulate and differentiate O₃ and PM_{2.5} pollution in key areas. We find two synoptic patterns are prone to lead to co-occurrence of O₃ and PM_{2.5} pollution: in the southern BTH and northern YRD 395 regions under Type 1 and the areas in the BTH and GZP under Type 2 are associated with the double





high level of O_3 and PM_{2.5}. About 55.6% of compound pollution occurrence days at all sites occurred under Type 1 while 33.0% under Type 2.

Type 1 weather pattern appears frequently in June, with a stable WPSH ridge line at about 22°N, and the warm and humid air brought by the WPSH reaches the area south of the Yangtze River, where a high temperature and high humidity Meiyu season is formed which suppresses the photochemical reaction of O₃ generation. Meanwhile, the north of China is controlled by a low-pressure trough with high temperatures and little rain. The hygroscopic growth of PM_{2.5} occurs in the corresponding area in front of the trough with a small amount of water vapor transported by the WPSH, causing compound pollution of O₃ and PM_{2.5} in the south of the BTH region and north of

405 the YRD region; particularly, unfavorable pollution diffusion conditions of a shallow BLH and low WS further exacerbate the severity of this compound pollution.

Under Type 2, the WPSH shifts northwards and retreats eastwards (sometimes retreats southwards and eastwards), with the meridional deepening of the East Asian major trough, and thus warm and humid airstreams are brought to the Northern China (e.g., the BTH region), gradually

- 410 elevating temperatures and humidity. Although positive RH anomaly promotes hygroscopic growth of the PM_{2.5}, water vapor leads a sink of ozone by contrast. As a result, the probability of double high level of O₃ and PM_{2.5} under Type 2 is less than Type 1, the extent of compound pollution in Type 2 is also narrowed. On the other hand, weak precipitation, shallow boundary layer and low wind speed tend to create favorable conditions for pollution maintenance.
- 415 In general, the location of the WPSH is tightly associated with O₃ pollution in eastern China, and the changes of meteorological conditions in different regions affected by the WPSH can induce significant regional differences in O₃ and PM_{2.5} pollution. More importantly, the effects of various large-scale weather circulation patterns on the O₃-PM_{2.5} compound pollution and their corresponding physical and chemical processes, have been clarified, which has important scientific
- 420 reference value in summer air-quality forecasts, as well as assessment and policy-making services.

Data availability

Hourly PM_{2.5}, NO₂, O₃, and O₃-8h data is published by the China Environmental Monitoring Station (<u>http://www.cnemc.cn/</u>). Surface meteorological data, such as Tmax, precipitation, WS and RH,





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 radiosonde data can be obtained from the China National Meteorological Information Center of the

 China Meteorological Administration (<u>http://data.cma.cn/site/index.html</u>). The NCEP/NCAR daily

 reanalysis
 dataset
 can
 be
 download
 from

 <u>https://psl.noaa.gov/data/gridded/data.ncep.reanalysis.html</u>.

Author contributions

L. Zong: Methodology, Data Curation, Formal Analysis, Writing- Original draft preparation, Results
 Discussion, Writing- Reviewing and Editing; Y. Yang: Conceptualization, Methodology, Formal
 Analysis, Results Discussion, Writing- Reviewing and Editing; M. Gao, H. Wang, P. Wang, L. Wang,
 H. Zhang, G. Ning, C. Liu, Y. Li, Z. Gao: Results Discussion, Comments, Writing- Reviewing and
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435 Competing interests

The authors declare that they have no conflict of interests

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	NEM				GZP				PRD				YRD				втн				Urban cluster N		
6~8	æ	7	6	6~8	80	7	6	6~8	œ	7	6	6~8	œ	7	6	6~8	œ	7	6		Month	_	
18889	2341	3422	13126	368	537	1879	1549	7735	977	1431	5327	28129	3593	5207	19329	9320	1188	1716	6416	days	Stas ×		
14.6%	4.4%	8.9%	17.9%	15.2%	28.5%	40.7%	42.9%	10.5%	25.4%	20.3%	5.2%	25.8%	25.5%	24.5%	26.2%	46.6%	18.9%	25.6%	57.3%	OLR	MD		
114.9	92.6	106.8	121.0	112.9	134.0	145.0	147.6	90.5	122.5	107.2	80.1	125.9	127.6	119.8	127.2	158.6	118.5	134.2	172.5	Con	DA O ₃ PM _{2.5}	Type1	
1.7%	1.0%	2.3%	1.7%	0.9%	1.6%	1.9%	0.4%	0.2%	1.1%	0.6%	0	4.0%	0.3%	5.3%	4.4%	16.3%	6.9%	21.2%	16.7%	OLR			
25.4	20.1	25.2	26.4	32.4	31.6	33.7	33.2	19.8	27.9	25.3	16.8	36.1	28.6	38.6	36.8	47.6	33.1	51.8	49.2	Con			
13101	5520	7338	243	2056	850	1168	38	5493	2316	3076	101	13754	19922	8459	11098	365	2805	3681	122	days	Stas ×		
13.4%	6.3%	17.0%	64.2%	43.1%	36.8%	47.4%	52.6%	13.5%	20.6%	7.6%	31.7%	17.4%	18.3%	22.0%	14.3%	54.2%	35.3%	54.2%	91.8%	OLR	MDA O ₃	Type2	
109.2	91.0	120.7	175.3	154.6	146.3	160.3	165.3	94.3	108.5	81.8	146.9	112.5	113.8	120.1	107.0	169.8	143.1	165.6	209.6	Con			
1.3%	1.3%	1.3%	0.4%	2.1%	1.6%	0	0	0.3%	0.6%	0.1%	0	1.7%	3.0%	0.6%	2.2%	18.8%	14.7%	22.5%	3.3%	OLR	PM2.5 OLR Con		
25.2	21.0	28.1	31.1	35.9	34.2	30.2	30.4	23.0	28.0	19.1	24.9	30.6	33.3	28.0	44.5	51.2	44.0	56.9	46.0	Con			
6124	3282	2722	120	981	530	432	19	2503	1376	1079	48	9309	4993	4135	181	3086	1671	1356	59	days	Stas ×		
5.8%	5.6%	5.7%	13.3%	26.8%	28.9%	24.3%	26.3%	14.3%	15.5%	13.3%	0	15.8%	16.9%	14.5%	12.7%	26.3%	18.9%	33.8%	62.7%	OLR	M	Type3	
96.4	98.3	93.0	123.3	138.9	143.1	133.3	150.8	98.2	103.4	92.4	.17	114.6	116.7	112.6	102.4	131.3	121.2	141.8	176.0	Con	DA O3		
0.8%	0.9%	0.7%	0.8%	1.5%	1.9%	1.2%	0	0.2%	0.3%	0	0	0.1%	0.0%	0.1%	0.6%	11.7%	10.1%	12.9%	32.2%	OLR	Ŧ		
22.4	23.2	20.8	37.6	34.7	37.2	31.7	34.7	21.8	24.0	19.3	13.2	24.0	23.7	24.0	32.0	44.8	42.5	46.7	67.1	Con	M _{2.5}		
3648	2507	1141	0	567	394	173	0	1454	993	461	0	5560	3817	1743	0	1845	1268	577	0	days	Stas ×		
10.1%	7.8%	15.1%	0	34.9%	38.6%	26.6%	0	14.8%	17.9%	8.0%	0	19.7%	14.7%	30.9%	0	35.5%	31.9%	43.5%	0	OLR	MDAO ₃	Type4	
105.5	97.8	122.4	0	151.6	154.9	144.1	0	102.1	111.4	82.1	0	122.5	114.8	139.5	0	147.3	144.3	153.7	0	Con			
0.2%	0.2%	0.2%	0	0.9%	1.3%	0	0	0.1%	0.1%	0	0	0.1%	0.1%	0.3%	0	6.9%	6.0%	8.8%	0	OLR	PM2.5		
21.0	19.5	24.3	0	35.8	39.9	26.4	•	22.8	25.1	17.9	0	26.7	25.1	30.0	0	42.8	39.7	49.5	0	Con			















Fig. 2. Time series of MDA8 O₃ pollution levels in key urban clusters.







Fig.3. Time series of PM_{2.5} pollution levels in key urban clusters.







Fig. 4. 850-hPa wind (vectors; see scale arrow at the bottom right in units of 5m/s) and 500hPa GH (contours; see scale bar at bottom in units of gpm) patterns based on objective classification (see text for details). Black box area indicates the area for classification and the white box area is for the area of eastern China, the number at the upper right corner of each panel indicates the frequency of the occurrence of each pattern type.







Fig. 5. Time series of synoptic circulation pattern.







Fig. 6. The MDA8 O₃ anomaly under four SWPs, where the sites marked with a '+' indicates

the Analysis of Variance passes the significance level of 0.05.







Fig. 7. The $PM_{2.5}$ anomaly under four SWPs, where the sites marked with a '+' indicates the

Analysis of Variance passes the significance level of 0.05.









Fig. 8. Daily variation of O₃ and PM_{2.5} under four SWPs in key urban clusters.







Fig. 9. The anomaly of Tmax (a–d), RH (e–h), and PF (i–l) under four SWPs, where the sites marked with a '+' indicates the Analysis of Variance passes the significance level of 0.05.







Fig. 10. The anomaly and significance of difference of BLH (a–d) and FLWD (e–h) under four SWPs, where the sites marked with a '+' indicates the Analysis of Variance passes the significance level of 0.05.







Fig. 11. NO_2 and $O_3\!/NO_2$ daily variation under four SWPs in key urban clusters.







Fig. 12. Schematic diagrams describing the relationships between the WPSH, four SWPs and

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summertime O₃ and PM_{2.5} pollution in various regions.