Dominant Patterns of Summer Ozone Pollution in Eastern China and Associated Atmospheric Circulations

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- Abstract. Surface ozone has been severe during summers in the eastern parts of China, damaging human's health and flora and 10 fauna. During 2015–2018, ground-level ozone pollution increased and intensified from south to north. In North China and Huanghuai region, the O₃ concentrations were highest. Two dominant patterns of summer ozone pollution were determined, i.e., a south-north covariant pattern and a south-north differential pattern. The anomalous atmospheric circulations composited for the first pattern manifested as a zonally enhanced East Asia deep trough and as a west Pacific subtropical high whose western ridge point shifted northward. The local hot, dry air and intense solar radiation enhanced the photochemical reactions
- 15 to elevate the O₃ pollution levels in North China and Huanghuai region. For the second pattern, the broad positive geopotential height anomalies at high latitudes significantly weakened cold air advection from the north, and those extending to North China resulted in locally high temperature near the surface. In a different manner, the west Pacific subtropical high transported sufficient water vapor to the Yangtze River Delta and resulted in locally adverse environment for the formation of surface ozone. In addition, the most dominant pattern in 2017 and 2018 was different from that in previous years, which is investigated as a new feature. Furthermore, the implications for the interannual differences in summer O₃ pollution have also proven to be
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meaningful.

Introduction 1.

Ozone occurs both in the stratosphere and at ground level. Stratospheric ozone forms a protective layer that shields us from the sun's harmful ultraviolet radiation. However, surface ozone is an air pollutant and has harmful effects on people and 25 on the environment, such as damaging human lungs (Day et al., 2017) and destroying agricultural crops and forest vegetation (Yue et al., 2017). Worldwide, severe ozone events are more frequent and stronger in China than those that have taken place in Japan, South Korea, Europe, and the United States (Lu et al. 2018). Due to their close relationship with anthropogenic emissions (Li et al., 2018), the high O₃ concentrations in China are mainly observed in urban regions, such as in North China (NC, 114 E-122.5 E, 36.5 N-42 N), the Yangtze River Delta (YRD, 117 E-122.5 E, 29.5 N-32.5 N) and the Pearl River 30 Delta (PRD, 111 E-115 E, 22 N-24.3 N) where rapid development has occurred in recent decades (Wang et al., 2017). An increase in surface ozone levels was found in China in 2016 and 2017 relative to 2013 and 2014 (Lu et al., 2018). The O_3 pollution levels in Beijing-Tianjin-Hebei (part of NC) were the most severe in China (Wang et al., 2006; Shi et al., 2015) and this situation has been getting worse. The O_3 concentrations in North China underwent a significant increase in the period of 2005–2015, with an average rate of 1.13 \pm 0.01 ppbv yr⁻¹ (Ma et al., 2016). Even on the highest mountain over NC, Mount Tai,

summer (June-July-August, JJA) O_3 increased significantly by 2.1 ppbv yr⁻¹ (Sun et al., 2016). The O_3 levels generally 35 presented increasing trends from 2012 to 2015 in the YRD (Tong et al., 2017), e.g., the O₃ concentrations in Shanghai (a mega-city) increased by 67% from 2006 to 2015 (Gao et al., 2017). In the PRD region, O_3 increased by 0.86 ppbv yr⁻¹ from 2006 to 2011 (Li et al., 2014). Severe ozone pollution is projected to increase in the future over eastern China (Wang et al.,

2013).

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- Although deep stratospheric intrusions may elevate surface ozone levels (Lin et al., 2015), the main source of surface ozone is the photochemical reactions between the oxides of nitrogen (NO_x) and volatile organic compounds (VOC), i.e., NO_x + $VOC = O_3$. The concentrations of NO_x and VOC are fundamental drivers impacting ozone production, and are sensitive to the regime of ozone formation, i.e., NOx-limited or VOC-limited (Jin and Holloway 2015). The changes in fine particulate matter are also a pervasive factor for the variation in ozone concentration. Employed Goddard Earth Observing System Chemical
- 45 Transport Model, Li et al (2018) found that rapid decreases in fine particulate matter levels significantly stimulated ozone production in NC by slowing down the aerosol sink of hydro-peroxy radicals. Furthermore, the meteorological conditions also influenced the ozone levels via modulation of the photochemical episodes (Yin et al., 2019; Lu et al., 2019). Intense solar radiation accelerated chemical O₃ production (Tong et al., 2017). A severe heat wave in the YRD contributed to high O₃ concentrations in 2013 (Pu et al., 2017). Winds had an impact on the O_3 and its precursors at downwind locations (Doherty et
- 50 al., 2013). Local meteorological influences are always related to specific large-scale atmospheric circulations. The changes in the East Asian summer monsoon led to 2-5% interannual variations in surface O₃ concentrations over central eastern China (Yang et al., 2014). Continental anticyclones created sunny and calm weather, which are favourable conditions for O_3 production in NC (Ding et al., 2013; Yin et al., 2019). Due to the associated transports of pollution from inland, tropical cyclones are often related to the evaluation of surface O_3 levels in the coastal areas of PRD (Ding et al., 2004). Basing on a case 55 study in 2014, further studies showed that a strong west Pacific subtropical high (WPSH) was unfavourable for the formation of O_3 in South China (Zhao and Wang, 2017), however the physical mechanisms to impact O_3 in North China was still not sufficiently explained.

Wang et al. (2017) reviewed the meteorological influences on ozone events, but the referenced findings were published mainly before 2010, when measurements in China were still scarce. Since 2015, O₃ measurements in eastern China were 60 steadily and widely implemented, but the O₃-weather studies mainly focused on meteorological elements (e.g. temperature, precipitation etc.) and several synoptic processes (Xu et al., 2017; Xiao et al., 2018; Pu et al., 2013). The dominant patterns of daily ozone in summer in east of China are still unclear. Actually, in our study, we found the most dominant pattern was different with that in Zhao and Wang (2017) and the dominant patterns also showed interannual variations. The findings of this study basically help to understand the varying features of daily surface ozone pollution in eastern China, their relationships with large-scale atmospheric circulations and the implications for the climate variability.

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2. Data sets and methods

Nationwide hourly O₃ concentration data since May 2014 are publicly available on http://beijingair.sinaapp.com/. Since the severe air pollution events in 2013, the air pollution issues gained more attentions from the Chinese government and society, which aided to start the extensive constructions of operational monitoring stations of atmospheric components and resulted in continuous increasing number of sites (Figure S1). The number of sites in eastern China (110°E–125°E, 22°N–42°N) was 677, 937, 937, 995 and 1007 from 2014 to 2018. It is obvious that the data in 2014 were deficient, while the observations were broadly distributed in eastern China and continuously achieved since 2015. Thus, the summer O₃ data from 2015 to 2018 were processed (e.g., unifying the sites and eliminating the missing value) and 868 sites in eastern China were employed here to reveal some new features of surface ozone pollutions and associated anomalous atmospheric circulations. Generally, severe air pollutions occurred more frequently in cites than in rural areas, therefore, the monitoring sites of atmospheric components mostly gathered around the urban areas, indicating the results of this study were more suitable to the urban O₃ pollution. The maximum daily average 8 h concentration of ozone (MDA8) is the maximum of the running 8 h mean O₃ concentration during an entire 24 hour day. According to the Technical Regulation on Ambient Air Quality Index of China (the Ministry of Environmental Protection of China, 2012), MDA8 is generally used to represent the daily O₃ conditions. The MDA8 \in [0, 100], (100, 160], (160, 215], (215, 265], (265, 800] µg/m³ corresponds to "Excellent", "Good", "Lightly polluted", "Moderately polluted", "Heavily polluted" levels of air quality in China.

The 2.5 °×2.5 °ERA-Interim data used here include the geopotential height (Z) at 850 and 500 hPa, zonal and meridional wind, relative humidity, vertical velocity, air temperature at from surface to 100 hPa, surface air temperature (SAT) and wind, downward solar radiation at the surface, low and medium cloud cover and precipitation (Dee et al., 2011). Because the maximum photochemical activity often occurred at afternoon (Wang et al., 2010), the daytime data were calculated by the 6-hourly reanalysis (including Z, wind, relative humidity, vertical velocity, air temperature and cloud cover) and 3-hourly reanalysis (precipitation and downward solar radiation) to composite the daytime atmospheric circulations and daytime meteorological conditions. Due to the different representative period of each element in ERA-Interim data, the daytime for Z, wind, relative humidity, vertical velocity, air temperature and cloud cover was from 05 a.m. to 05 p.m (Beijing Time; 21 p.m.–09 a. m. UTC), while it is from 08 a.m. to 08 p.m. (Beijing Time; 00 a.m. to 00 p.m. UTC) for precipitation and downward solar radiation.

The empirical orthogonal function (EOF) analysis is a widely used statistical method in meteorology to reconstruct the original variables into several irrelevant patterns (Wilks, 2011). The EOF analysis, applied to the daily anomalies (MDA8 anomalies at 868 stations in this study), extracted the relatively change features of the original data on the daily time-scale. The orthogonal modes included spatial and temporal coefficients, and contained information of some proportion (variance contributions) from the original fields. Significance test must be execute to confirm whether the decomposed patterns had physical meanings. In this study, we used the test method form North et al., (1982). That is, if the eigenvalue (λ) satisfied the condition as $\lambda_i - \lambda_{i+1} \ge \lambda_i (2/n)^{1/2}$, the eigenvalue λ_i was significantly separated. We performed this significance test on the selected patterns from EOF decompositions, and confirmed that these dominant patterns in this study were all significant. The aforementioned EOF analysis programs were finished by the NCAR Command Language.

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3. Variations and dominant patterns

During 2015–2018, summer surface ozone pollution was severe in China, especially in the economically developed regions. Spatially, the JJA mean MDA8 increased from south to north in eastern China (Figure 1a). To the south of 28°N (i.e., South China), the mean MDA8 was mostly lower than 100 µg/m³ and the ozone pollution was obviously lower than that in 105 North China and in the Huanghuai area (NCH). It is notable that, although the values of MDA8 in the PRD were not as large as those in NCH, they were relatively higher than those in the surrounding areas. The mean MDA8 was above $110 \,\mu\text{g/m}^3$ to the north of 32°N (i.e., the NCH area), and thereinto, the large values of MDA8 centred on the Beijing-Tianjin-Hebei region and in western Shandong province exceeded 150 µg/m³. In the transitional zone, i.e., between 28°N and 32°N, the MDA8 varied from 100 µg/m³ to 120 µg/m³. Surface O₃ pollution was closely linked to the anthropogenic emissions that dispersed and 110 concentrated in the large cities (Fu et al., 2012), which was similar to the haze pollution (Yin et al., 2015). In the YRD and PRD, high levels of MDA8 were scattered around the large cities. However, the high-level O_3 values in the NCH region were contiguous, indicating extensively severe surface O₃ pollution both in large and small cities (Figure 1). Furthermore, the maximum values of MDA8 for four summers were extracted to evaluate the severest levels of O₃ pollution (Figure 1b). To the north of 30°N, the maximum MDA8 at most sites was above $265\mu g/m^3$, indicating that the levels of O₃ pollution had exceeded the threshold of heavily O₃ pollution in China (The Ministry of Environmental Protection of China, 2012).

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Ten cities, with severe O₃ pollutions, were chosen to investigate the temporal variations, including Beijing (capital of China), Tangshan, Tianjin near the capital city, Shijiazhuang, Weifang and Taiyuan in the south of NCH, Nanjing and Shanghai in YRD, Guangdong and Zhongshan in PRD (Figure S2). These cities had large populations and were with severe O₃ pollutions. In Beijing, Tianjin and Tangshan, the MDA8 values were nearly above 100 µg/m³ and frequently exceeded 215 μ g/m³ (Figure 2a). The percentage of non-O₃-polluted days (<100 μ g/m³) and moderate O₃-polluted days (>215 μ g/m³) were 14.9% and 15.5% for the mean MDA8 of these three cities. The former percentage indicated that more than $85\% O_3$

concentrations exceeded the health threshold, and the later meant, in more than 15% of summer days, O_3 concentrations moderately damaged human health in the Beijing-Tianjin-Hebei region. The maximum MDA8 in the north of Hebei province (e.g. Tangshan in Figure 2a) and in eastern Shandong province (e.g. Weifang in Figure 2b) even exceeded 320 μ g/m³, which

- badly injured the health of local citizens. In Shijiazhuang, Weifang and Taiyuan, the MDA8 levels were lower than those in Beijing and Tianjin during 2015–2016, but dramatically increased to levels comparable to those of Beijing and Tianjin in 2017 and 2018 (Figure 2b). In Nanjing and Shanghai, the MDA8 did not show a clear increasing trend (Figure 2c). Similar to the distribution of the mean MDA8, the maximum MDA8 to the south of 30°N was lower by comparison. Although approximately 60% of summer days were non-O₃-polluted in the cities of Guangzhou and Zhongshan (Figure 2d), severe O₃ pollution also
- 130 occurred in the PRD (Figure 1b).

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Considering the characteristics of the observed MDA8 mentioned above, EOF approach was used to explore the dominant patterns of summer ozone pollution in eastern China (Figure 3). The percentages of variance contribution for the first three patterns were 21.5%, 15.5% and 8%. The significance test of the EOF eigenvalues confirmed that the first three patterns were significantly separated. Approximately 37% of the variability in the original data was contained in the first two patterns, therefore, they were defined as the dominant patterns of surface ozone pollution on the daily time-scale. In the first EOF pattern (PAT1), the observed MDA8 at different sites changed similarly and the centre of variation was located in the NCH area (Figure 3a). The time series of EOF1 showed that the ozone pollution during 2017–2018 was more serious than that in 2015 and 2016 (Figure 3b). Differently, the second EOF pattern (PAT2), showed notable south-north difference, with centres in the NC and YRD regions (Figure 3c). The time coefficient of PAT2 also did not show an obvious increasing trend (Figure 3d). The positive (P) and negative (N) phases of PAT1 (PAT1P, PAT1N) and PAT2 (PAT2P, PAT2N) are defined by the events that are greater than one standard deviation and less than $-1 \times$ one standard deviation, respectively (Figure 3b, 3d).

Figure 3 illustrates the EOF results for the dominant patterns of surface ozone, while Figure 4 showed the MDA8 composites break down into the positive and negative phases. The ozone concentrations for the PAT1P classification (Figure 4a) were generally greater than those for PAT1N (Figure 4b). Most of the MDA8 values in the NCH region were >160 µg/m³
and <120 µg/m³ for PAT1P and PAT1N, respectively. For the second pattern, the PAT2P appeared as a diminishing pattern from the north to the south (Figure 4c), however, there was severe ozone pollution in the YRD and PRD under PAT2N conditions (Figure 4d). Therefore, the centres of O₃ variation were NCH for the PAT1, and NC and the YRD for the PAT2.

4. Associated atmospheric circulations

In eastern China, the economic productions and human actives steadily developed in the recent four years and the emissions of ozone precursors were reasonably supposed to be relatively stable on the daily time-scale. Differently, the daily variations in MDA8 were evidently saw in Figure 2. Therefore, the impacts of daily meteorological conditions significantly contributed to the domain patterns of daily O_3 concentrations and their variations. Anomalous daytime atmospheric circulations associated with PAT1 (PAT1P composite minus PAT1N composite) and PAT2 (PAT2P composite minus PAT2N composite) were shown Figure 5–6. For example, the mean of the atmospheric circulations associated PAT1P (PAT1N) were

155 firstly computed, and then the differences between PAT1P composites and PAT1N composites were calculated as the anomalous daytime atmospheric circulations associated with PAT1. For the first pattern, the largest O₃ differences between the PAT1P and PAT1N was within the NCH region (Figure 4a, b). The correlation coefficient between the time series of PAT1 and the NCH-averaged MDA8 was 0.97 (Table 1). Thus, the effects of the anomalous atmospheric circulations mainly acted on the photochemical reactions near the surface in NCH. There were negative Z850 anomalies over the Ural Mountains. Over the broad region from eastern Eurasia to the north Pacific, the anomalous atmospheric circulations were located zonally, i.e., positive Z850 on the tropical zone, cyclonic anomalies at the mid to high latitudes and positive anomalies on the polar region (Figure 5a). The East Asia deep trough was enhanced and extended to northeast China and Japan. The intensity of the East Asia deep trough (i.e., the negative area-averaged Z850) positively correlated with the time series of PAT1 (Table 1) with a

correlation coefficient of 0.28 (above the 99% confidence level). In accordance with the deep positive height anomalies to the

north of Lake Baikal (centring at 107 °E, 53.5 °N), which also extended southward to the edge of the Tibetan Plateau, cold air

was transported to the lower latitudes, but did not arrive at the NCH region (Figure 5a).

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Influenced by the enhanced East Asia deep trough, the main body of WPSH shifted southward (compared to its climate status in summer). The location of WPSH ($Z500_{(125^{0}E, 20^{0}N)} - Z500_{(125^{0}E, 30^{0}N)}$) also showed a positive correlation with the time series of PAT1 (R=0.39, Table 1). However, the western ridge point of WPSH was northward and westward than normal (being indicated by $Z500_{(110^{0}E, 30^{0}N)}$), and occupied the NCH area, which was significant with the time series of PAT1 (R=0.24, above the 99% confidence level). Although the local anomalous anticyclone over the east of China seemingly delivered water vapor to North China (Figure 5b), the channel of moisture was already cut off in the ocean at low latitudes by the positive and zonal anomalies in the tropical regions (Figure 5a) and resulted in a dry environment in NCH from surface to 400 hPa (Figure 5c). Furthermore, the associated descending motions (Figure 5c) not only corresponded to the warmer surface air temperature (Figure 5a), but also suppressed the development of convective activity (indicating by less low and medium cloud, Figure 5d). The correlation coefficients between the time series of PAT1 and NCH-averaged precipitation, SAT, and downward solar radiation at surface were -0.44, 0.14 and 0.45, respectively, all of which exceeded the 99% significance test (Table 1). The large-scale atmospheric circulations led to days with high temperatures near the surface (Figure 5a), less precipitation (Figure 5b), a dry environment (Figure 5c) and intense solar radiation (Figure 5d), which substantially enhanced respectively).



For PAT2, largest O₃ differences (PAT2P composite minus PAT2N composite) were observed in the NC and YRD regions (Figure 3c, Figure 4c, d). The correlation coefficient between the time series of PAT2 and the MDA8 difference

between NC and the YRD was 0.77 (Table 1). The impacts of atmospheric circulations on the photochemical reactions in the above two areas are analysed in Figure 6. Due to the broad positive Z500 anomalies at the high latitudes of Eurasia, the

- 185 subjacent surface air temperatures significantly increased, indicating weak cold air advection from the north (Figure 6a). Moreover, there were positive Z500 anomalies from the Chukchi Peninsula to Northeast China. In summer, anomalous anticyclonic circulations at the mid and high latitudes generally led to significantly positive SAT anomalies (Figure 6a). The East Asia deep trough was stronger (R=0.3), but was limited to the east of Japan.
- Extruded by the East Asia deep trough and cyclonic anomalies from the Siberian plains to the YRD, the WPSH moved 190 southward and exhibited southwest-northeast orientation (Figure 6a). The location of WPSH $(Z500_{(110^{0}E - 20^{0}N)} Z500_{(110^{0}E, 30^{0}N)}$) was positively correlated with the time series of PAT2 (R=0.32, Table 1). The southwest-northeast distribution of WPSH aided water vapor transportation to the YRD region (Figure 6b-c). Combined with significant upward air flow (Figure 6c), more clouds formed at the medium and low levels (Figure 6d) and precipitation was enhanced in the YRD region (Figure 6b). A moist-cool environment, weak solar radiation and obvious wet deposition reduced the ozone 195 concentration in the YRD region. On the other hand, sinking motion (Figure 6c) and less cold air advection from the north (Figure 6a) both resulted in a temperature increase in NC (Figure 6a). There was divergence of water vapor and less cloud cover over NC, resulting in dry, hot and sunny weather (Figure 6b, d). Under such meteorological conditions, the generation of surface O₃ was accelerated, and thus, higher MDA8 was observed in NC. The differences in precipitation, SAT, and downward solar radiation at the surface between the NC and YRD regions were calculated and their correlation coefficients with the time 200 series of PAT2 were -0.46, 0.18 and 0.62, respectively (Table 1). The significant correlations indicated that the differences in meteorological conditions between NC and YRD regions, associated with the aforementioned anomalous atmospheric circulations, largely contributed to O₃ PAT2.

5. Signals for interannual variability

The O₃ pollutions in NCH and YRD were severe over the past four years, reflected in both the O₃-polluted areas and in the O₃ concentration (Figure 1). The number of sites with maximum MDA8 > 265 μ g/m³ in NCH (YRD) was 94 (35), 55 (22), 180 (58), 160 (46) from 2015 to 2018 (Figure 7). The summer mean MDA8 in the PRD was not as high as that in NC and the YRD (Figure 1a), but maximum O₃ concentration exceeded 265 μ g/m³ could also be observed in certain large cities of PRD in each year (Figure 7). Additionally, the observed summer MDA8 anomalies in eastern China presented evident interannual differences (Figure 7). The dominant spatial patterns of MDA8 anomalies in each year were also different (Figure 8). Although the relative variance contributions of the spatial coefficients varied, the first two EOF patterns of MDA8 were always PAT1 and PAT2 in different years, indicating that the extracted dominant patterns were reliable and steady. Sorting by the variance contribution, the dominant patterns were PAT2 and PAT1 in 2015 and 2016 (Figure 8a–d), however, they are PAT1 and PAT2 in the two subsequent years (Figure 8e–h). The first EOF pattern in 2014 revealed by Zhao and Wang (2017) was similar with PAT2, however the most dominant pattern changed to PAT1 in the latest two years (2017 and 2018).

A question raised here is whether the aforementioned composited signals of atmospheric circulations could provide implications for the climate variability of the summer O₃ pollution in eastern China. In 2016 and 2018, the variance contribution of the first pattern was almost twice that of the second pattern, and thus, these two years were selected as typical years whose varied patterns were clearly separated. The dominant pattern of 2016 was PAT2 (explaining approximately 24% of the variance, Figure 8c), while that in 2018 changed as PAT1, with nearly 34% variance contributions (Figure 8g). In 2016, the MDA8 values in NC and the YRD were nearly out of phase (Figure 9a), and the correlation coefficient between them was –0.28 (above the 99% confidence level). Differently, this correlation coefficient was 0.43 in 2018 (Figure 9b), indicating similar change features between the MDA8 levels of NC and the YRD.

The MDA8 anomalies in 2016 were negative in NC, but positive in the YRD and PRD (Figure 7b), which was the opposite pattern of PAT2. The interannual anomalies of atmospheric circulations in 2016, with respect to the mean of 225 2015–2018 (Figure 10), were almost opposite to the anomalous atmospheric circulations associated with PAT2 (Figure 6). There were positive Z500 anomalies over the north Pacific at the mid to high latitudes (Figure 10a). These positive anomalies not only indicated a weaker East Asia deep trough but also induced a shallow trough from the Chukchi Peninsula to Northeast China. Together with the stronger high ridge over the Ural Mountains, the cold air was transported to the mid latitudes, resulting in a lower SAT in North China (Figure 10a). The western segment of the WPSH was stronger and moved northward, 230 which occupied the YRD and south China (Figure 10a) and brought moist air flows to North China (Figure 10b). Sufficient moisture formed more low to mid-level clouds, causing a decrease in solar radiation reaching the ground in NC (Figure 10d). Associated with the extending WPSH, sinking motions resulted in a hot, dry air mass near the surface in the YRD region (Figure 10b-c). In addition, decreased cloud cover did not effectively reflect solar radiation, which was an essential condition for the enhancement of the photochemical reactions. Therefore, at the interannual time scale, the atmospheric anomalies, 235 opposite to those shown in Figure 6, also played important roles on the spatial pattern of MDA8 anomalies. That is, the atmospheric circulations in 2016 accelerated the formation of surface O_3 in the south of YRD, but weakened the summer O_3 pollution in NC.

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The MDA8 anomalies were mostly positive in the east of China in 2018 (Figure 7d). "-+-" pattern of Z850 anomalies were located over the Ural Mountains and to the north of Lake Baikal and the Aleutian Islands (Figure 11a), which was consistent with the anomalous patterns in Figure 5. The East Asia deep trough shifted northward than the mean status during 2015–2018, and meanwhile, the western ridge point of WPSH also shifted northward, resulting in a higher SAT in the east of China (Figure 11a) and accelerating the photochemical conversion for elevating the surface ozone concentration. The local anomalous anti-cyclone over the NCH and the Japan Sea also existed in the interannual signals, which induced the divergence

of water vapor in southeast China (Figure 11b). Due to the lack of moisture, it was difficult to form cloud cover, and more solar

radiation directly reached the ground (Figure 11d). The large-scale atmospheric circulations led to high temperatures near the surface in eastern China, and to a dry and sunny environment in the YRD and South China. Thus, under such weather conditions, positive MDA8 anomalies were observed in 2018. Although the signals of global warming had impacts on the anomalies of atmospheric circulations, the key features could also be found when the anomalies were viewed with respect to the climate mean during 1979–2018 (Figure S3–S4). In addition, if the variance contributions of the first two EOF patterns were not significantly different, i.e., in 2015 and 2017, the interannual signals of atmospheric circulations also showed integrated characteristics of PAT1 and PAT2 (Figure S5–S6).

6. Conclusions and discussions

During the recent four years, ground-level ozone pollution became the major air challenge in the summers in the east of China. The highest O₃ concentrations were observed in North China and in the Huanghuai region, which are located north of 32°N. The O₃-contaminated air occurred for 85% of summer days in Beijing and Tianjin. In the south, the surface O₃ pollution was also severe both in the Yangtze and Pearl River delta regions. Meteorological conditions had significant impacts on the evident daily fluctuation of MDA8. To reveal their detailed relationships, the dominant patterns of summer ozone pollution and associated atmospheric circulations were analysed in this study.

The MDA8 of the first prominent pattern changed synergistically in the east of China, especially in North China and in the Huanghuai region. An enhanced East Asia deep trough and west Pacific subtropical high were zonally distributed and prevented the northward transportation of moisture. The northward-shifted western ridge point of the west Pacific subtropical high accelerated the photochemical reactions via hot-dry air and intense solar radiation. The second pattern of ozone pollution showed remarkable south-north differences. Broad positive geopotential height anomalies at the high latitudes significantly increased the surface air temperature and thus decreased cold air advection from the north. These positive anomalies also extended to North China and resulted in locally warmer air near the surface. On the other hand, the southwest-northeast oriented west Pacific subtropical high transported sufficient water vapor to the Yangtze River Delta. Consequently, a local moist-cool environment, without intense sunlight, reduced the formation of surface ozone.

In this study, we mainly emphasized the contribution of the meteorological impacts and assumed the emissions of ozone precursors were relatively stable on the daily time-scale. There is no doubt that the human activities were the fundamental driver of air pollution even on the daily time-scale. However, the daily emission data were difficult to be acquired, thus the joint effects of the daily meteorological conditions and anthropogenic emissions needed to be discussed in future work. The interannual differences in summer O₃ pollution were also discussed and the composited signals of atmospheric circulations and weather conditions proved to have meaningful implications for climate variability. Lu et al., (2019) found that the observed 2017 surface ozone increases relative to 2016 in China are largely due to hotter and drier weather conditions, while changes in

- 275 domestic anthropogenic emissions alone would have led to ozone decreases in 2017. Although the simultaneous large-scale atmospheric circulations were diagnosed on an interannual scale, their possible preceding climate drivers, e.g., sea ice, and sea surface temperature, were still unclear. The research related to climate variability has always needed long-term data. To get around the problem of the data time span, Yin et al (2019) developed an ozone weather index using data from 1979 to 2017 and demonstrated the contributions of Arctic sea ice in May to O₃ pollution in North China. In addition, the dominant patterns of
- 280 ozone concentrations were also decomposed with the observed data from 2015 to 2018. With the increase in O₃ observations, increasingly reliable dominant patterns and more features might be revealed in the future. At present, the fine particulate matter decreased in the summers in eastern China (Li et al. 2018); however ozone production was significantly enhanced. Thus, attentions to surface pollution should be strengthened and the weather-climate component should be taken into account when making decisions for control measures.

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Author contribution

290 ZY and HW designed the research. BC and ZY performed most of the Figures and analysis. ZY prepared the paper with contributions from all co-authors.

Competing interests

The authors declare that they have no conflict of interest.

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Figures captions

Table 1. Correlation coefficients between the time series of PAT1 (PAT2) and the key indices of atmospheric circulations and meteorological conditions. "**" and "*" indicate that the correlation coefficients were above the 99% and 95% confidence level, respectively.

Zhao, Z. J., Wang, Y. X.: Influence of the west pacific subtropical high on surface ozone daily variability in summertime over eastern china, Atmospheric Environment, 170, 197–204, https://doi.org/10.1016/j.atmosenv.2017.09.024, 2017.

Figure 1. Distribution of the (a) mean values and (b) maximum values of MDA8 (Unit: $\mu g/m^3$) at the observation sites in summer from 2015 to 2018.

Figure 2. Variations in MDA8 (Unit: $\mu g/m^3$) of polluted cities from 2015 to 2018, including (a) Beijing, Tianjin and Tangshan; (b) Taiyuan, Weifang and Shijiazhuang; (c) Shanghai and Nanjing; and (d) Zhongshan and Guangzhou. The cites in panels (a)-(d) were located from north to south. The horizontal dash lines indicated the value of 100 $\mu g/m^3$ and 215 $\mu g/m^3$.

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Figure 4. Composites of the MDA8 (Unit: $\mu g/m^3$) for PAT1 (a, b) and PAT2 (c, d) in summer from 2015 to 2018. Panels (a) and (c) were composited when the time coefficient of EOF1 and EOF2 was greater than one standard deviation, while panels (b) and (d) were composited when the time coefficient was less than -1×0^{-1} some standard deviation.

- Figure 5. Differences of the daytime atmospheric circulations (i.e., PAT1P minus PAT1N). (a) Geopotential height at 850
 hPa (Unit: 10gpm, contours) and surface air temperature (Unit: K, shading), (b) water vapor flux (Unit: kg*m/(kg*s)) at 850
 hPa (arrows) and precipitation (Unit: mm, shading), (c) 100°E–120°E mean wind (Unit: m/s, arrows) and relative humidity (Unit: %, shading), (d) downward solar radiation at the surface (Unit:10⁷ J/m², shading) and the sum of low and medium cloud cover (Unit: 1, contours). The white dots indicate that the shading was above the 95% confidence level. The green boxes in panels (a), (b) and (d) show the NCH region, and the black box in panel (a) indicates the location of the East Asia
- trough. The purple triangles in panel (a) indicated the data used to calculate the WPSH₁, while the red triangle represented the west ridge point of WPSH.

Figure 6. Differences of the daytime atmospheric circulations (i.e., PAT2P minus PAT2N). (a) Geopotential height at 500 hPa (Unit: 10gpm, contours) and surface air temperature (Unit: K, shading), (b) water vapor flux (Unit: kg*m/(kg*s)) at 850 hPa (arrows) and precipitation (Unit: mm, shading), (c) 100°E–120°E mean wind (Unit: m/s, arrows) and relative humidity

- 420 (Unit: %, shading), (d) downward solar radiation at the surface (Unit: 10⁷J/m², shading) and the sum of low and medium cloud cover (Unit: 1, contours). The white dots indicate that the shading was above the 95% confidence level. The green boxes in panel (a), (b) and (d) are the NC and YRD regions, and the black box in panel (a) indicates the location of the East Asia trough. The purple triangles in panel (a) indicated the data used to calculate the WPSH₂.
- **Figure 7.** Anomalies of the summer mean MDA8 (Unit: $\mu g/m^3$) in 2015 (a), 2016 (b), 2017 (c) and 2018 (d), relative to the mean during 2015–2018. The black pluses indicate that the maximum MDA8 was larger than 265 $\mu g/m^3$.
- **Figure 8**. The first (a, c, e, g) and second (b, d, f, h) EOF spatial patterns of MDA8 in summer in 2015 (a, b), 2016 (c, d), 2017, (e, f) and 2018(g, h). The percentage number in panels (a, c, e, g) and (b, d, f, h) are the variance contributions of the first and second EOF mode.

Figure 9. Variations in the MDA8 (Unit: µg/m³) of NC (black) and the YRD (blue) in 2016 (a) and 2018 (b).

- Figure 10. Anomalies of summer mean daytime atmospheric circulations in 2016, with respect to the mean during 2015–2018. (a) Geopotential height at 500 hPa (Unit: 10gpm, contours) and surface air temperature (Unit: K, shading), (b) water vapor flux (Unit: kg*m/(kg*s)) at 850 hPa (arrows) and precipitation (Unit: 0.1mm, shading), (c) 100°E–120°E mean wind (Unit: m/s, arrows) and relative humidity (Unit: %, shading), (d) downward solar radiation at the surface (Unit: 10⁴J/m², shading) and the sum of low and medium cloud cover (Unit: 1, contours). The green boxes in panel (a), (b) and (d) are the NC and YRD regions. The white dots indicate that the shading was above the 95% confidence level.
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Table 1. Correlation coefficients between the time series of PAT1 (PAT2) and the key indices of atmospheric circulations and meteorological conditions. "**" and "*" indicate that the correlation coefficients were above the 99% and 95% confidence level, respectively.

PAT1	MDA81	EAT ₁	WPSH ₁	Pre ₁	SAT ₁	SSR ₁
	0.97^{**}	0.28^{**}	0.39**	-0.44^{**}	0.14^{**}	0.64**
PAT2	MDA8 ₂	EAT ₂	WPSH ₂	Pre ₂	SAT ₂	SSR ₂
_	0.77^{**}	0.30**	0.32**	-0.49^{**}	0.18^{**}	0.65**

445 MDA8₁ is the NCH-area averaged MDA8, while the MDA8₂ is the MDA8 difference between NC and YRD. EAT₁ and EAT₂ indicate the intensity of the East Asia deep trough and were calculated as the mean -Z850 shown in the black boxes in Figure 5 and Figure 6, respectively. WPSH₁ ($Z500_{(125^{0}E, 20^{0}N)} - Z500_{(125^{0}E, 30^{0}N)}$) and WPSH₂ ($Z500_{(110^{0}E, 20^{0}N)} - Z500_{(125^{0}E, 30^{0}N)}$) $Z500_{(110^{0}E, 30^{0}N)}$) represents the location of WPSH. Pre₁, SAT₁ and SSR₁ were calculated as the NCH-area averaged precipitation, SAT and downward solar radiation at the surface, respectively. Pre2, SAT2 and SSR2 are the differences in the NC- and YRD-area averaged precipitation, SAT and downward solar radiation at the surface, respectively.



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485

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Figure 6. Differences of the daytime atmospheric circulations (i.e., PAT2P minus PAT2N). (a) Geopotential height at 500 hPa (Unit: 10gpm, contours) and surface air temperature (Unit: K, shading), (b) water vapor flux (Unit: kg*m/(kg*s)) at 850 hPa (arrows) and precipitation (Unit: mm, shading), (c) 100°E–120°E mean wind (Unit: m/s, arrows) and relative humidity (Unit: %, shading), (d) downward solar radiation at the surface (Unit: 10⁷J/m², shading) and the sum of low and medium cloud cover (Unit: 1, contours). The white dots indicate that the shading was above the 95% confidence level. The green boxes in panel (a), (b) and (d) are the NC and YRD regions, and the black box in panel (a) indicates the location of the East Asia trough. The purple triangles in panel (a) indicated the data used to calculate the WPSH₂.



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Figure 11. Anomalies of summer mean daytime atmospheric circulations in 2018 with respect to the mean during 2015–2018. (a) Geopotential height at 850 hPa (Unit: 10gpm, contours) and surface air temperature (Unit: K, shading), (b) water vapor flux (Unit: kg*m/(kg*s)) at 850 hPa (arrows) and precipitation (Unit: 0.1mm, shading), (c) 100°E–120°E mean wind (Unit: m/s, arrows) and relative humidity (Unit: %, shading), (d) downward solar radiation at the surface (Unit: 10⁴J/m², shading) and the sum of low and medium cloud cover (Unit: 1, contours). The green boxes in panels (a), (b) and (d) show the NGU radian.

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