https://doi.org/10.5194/acp-2020-554
Preprint. Discussion started: 21 July 2020

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Ozone affected by a succession of four landfall typhoons in the Yangtze River Delta, China: major processes and health impacts

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Abstract: Landfall typhoon can significantly affect O_3 in the Yangtze River Delta (YRD) region. In this study, we investigate a unique case characterized by two multiday regional O_3 pollution episodes related to four successive landfall typhoons in the summer of 2018 in the YRD. The results show that O_3 pollution episodes mainly occurred during the period from the end of typhoon and the arrival of the next typhoon. The moment that typhoon reached the 24-h warning line and the last moment of typhoon activity in the mainland China can be roughly regarded as time nodes. Meanwhile, the variations of O_3 was related to the track, duration and landing intensity of the typhoons. The impact of typhoon on O_3 was like a wave superimposed on the background of high O_3 concentration in the YRD in summer. When typhoon was near the 24-h warning line before it landed the coast line of the YRD, the prevailing wind originally from the ocean changed to from the inland, and transported lots of precursors from the polluted areas to the YRD. With typhoon, the low temperature, strong upward airflows, more precipitation and wild wind prevented high O_3 episodes.

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After typhoon, the air below the 700 hPa atmospheric layer was warm and dry, which was conductive to the formation of O_3 from the abundance of precursors. It is note-worthy that O_3 is mainly generated in the middle of boundary layer (~ 1000 m), and then transported to the surface by downward airflows or turbulences. Moreover, O_3 can be accumulated and trapped on the ground due to the poor diffusion conditions because the vertical diffusion and horizontal diffusion were suppressed by downward airflows and light wind, respectively. The premature mortalities attributed to O_3 exposure in the YRD during the study period is 194.0, more than the casualties caused directly by the typhoons. This work enhances our understanding of how landfall typhoons affect O_3 in the YRD, which can be helpful to forecast the O_3 pollution synthetically impacted by the subtropical high and typhoon.

Key Words: ozone; landfall typhoon; the Yangtze River Delta region;

1 Introduction

The tropospheric ozone (O_3), which is formed by a series of complex photochemical reactions between volatile organic compounds (VOCs) and nitrogen oxides (NO_x=NO+NO_2) in combination with sunlight (Chameides and Walker, 1973; Xie et al., 2014), has received continuous attention due to its negative impact on air quality (Chan and Yao, 2008; Monks et al., 2015), human health (Jerrett et al., 2009), climate (Allen et al., 2012; IPCC, 2014) and biosphere (Dingenen et al., 2009). Research on urban O_3 pollution can date back to the early 1950s, beginning with the Los Angeles smog. In China, the photochemical smog, which is characterized by high level of O_3, was first discovered in Xigu district of Lanzhou in 1970s (Tang et al., 1989). However, with the key atmospheric environmental problem was coal-smoke pollution (such as acid rain) at that time (Wang et al., 2019), little systematic research and coordinated O_3 monitoring were performed in China until the mid-2000s (Wang et al., 2017).

Since the beginning of 21st century, the complex air pollution, which is dominated by fine particulate matter ($PM_{2.5}$, particles of 2.5 microns or less in aerodynamic diameter) and surface O_3, has been ingrained in the megacities of China (Chan and Yao, 2008; Jin et al., 2016; Kan et al., 2012). Air pollution has evolved into a political and economic concern in China. Due to drastic air pollution control since 2013, particle pollution has been greatly reduced, appearing a significantly decrease in sulfur dioxide (SO_2), NO_x and $PM_{2.5}$. However, the concentrations of O_3 and VOCs had

https://doi.org/10.5194/acp-2020-554
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an increase from 2013 to 2017 (Li et al., 2017), suggesting that more attention should be paid to controlling O_3 and VOCs in the future. Overall, the causes of air pollution and the control policies are still a major challenge in China, especially in understanding the sources, transport and dispersion processes, and chemical formation mechanisms of O_3 and its precursors (Ding et al., 2016; Guo et al., 2014; Huang et al., 2014).

Typhoon (tropical cyclone, TC) is one of the most important factors of natural disasters in East Asia. Out of the total provinces in China, 10 coastal and 6 island provinces are affected by typhoon induced disasters, with more than 250 million lives are affected (Liu et al., 2009). The average number of typhoons making landfall in China is 9 each year, and those typhoons usually inflict vast losses in human life and property due to the accompanied strong wind, torrential rains and huge storm surges (Zhang et al., 2009; Zhao et al., 2012). Because of the long lifetime and tremendous energy, typhoon has a significantly influence on local atmospheric conditions, and thereby can affect surface O_3 concentration through advection, diffusion, deposition and other processes. The impact of typhoon on O_3 has attracted extensive attention in recent years (Deng et al., 2019; Huang et al., 2005; Shu et al., 2016; Wang and Kwok, 2002; Wei et al., 2016; Yang et al., 2012). For example, Deng et al. (2019) reported that high O_3 and high aerosol concentrations (double high episodes) are likely to occur when the PRD is under the control of typhoon periphery and subtropical high with strong downdrafts. Previous studies were mainly in the southern China (including Hong Kong and Taiwan), where are frequently affected by typhoons. However, research on the impact of landfall typhoons on O_3 is still limited.

The Yangtze River Delta (YRD) region, being one of the most developed and densely populated regions in China, is located on the western coast of the Pacific Ocean. With 3.7% of the area and 16.0% of the population of China, the YRD contributed over 20% of the national total Gross Domestic Product (GDP) in 2019. Due to the rapid economic development and high energy consumption, this region is suffering from intense air pollution (Ding et al., 2013; Li et al., 2019; Wang et al., 2015; Xie et al., 2016). In 2017, the 90th percentile of the maximum daily 8-hour average (MDA8) O_3 concentration was 170 μg m^{-3}, and 16 of the 26 cities (Figure 1b) in the YRD failed to meet national standard (http://www.cnemc.cn/jcbg/zghjzkgb/201905/t20190529_704755.html). Therefore, it is an urgent task to investigate the spatiotemporal characteristic of O_3 as well as its formation mechanism in the YRD. Influenced by monsoon weather, the warm and stagnation

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conditions play an important role in the occurrence of high-level O_3 in summer (Li et al., 2018; Liao et al., 2015; Lu et al., 2018; Zhao et al., 2010). Synoptic weather systems, such as typhoon and cold front, also have significant effects on O_3 in the YRD (Hu et al., 2013; Shu et al., 2016). This work aims to reveal the main processes of landfall typhoon affecting surface O_3 in the YRD, hoping to fill the knowledge gap and be helpful for making reasonable pollution control measures.

In this study, we report an outstanding case observed in the YRD during the period from 16 July to 25 August, during which multiday episode of high O_3 occurred and was related to four successive landfall typhoons. Base on the monitoring data and numerical simulation, we further explore the impact of landfall typhoons on O_3 in the YRD, including the major processes and health impacts. The following part of this paper is structured as the follows: Section 2 gives a brief description of monitoring data, the analysis methods, and model configurations. The results as well as the discussions are detailed in section 3. Section 4 summarizes the main conclusions.

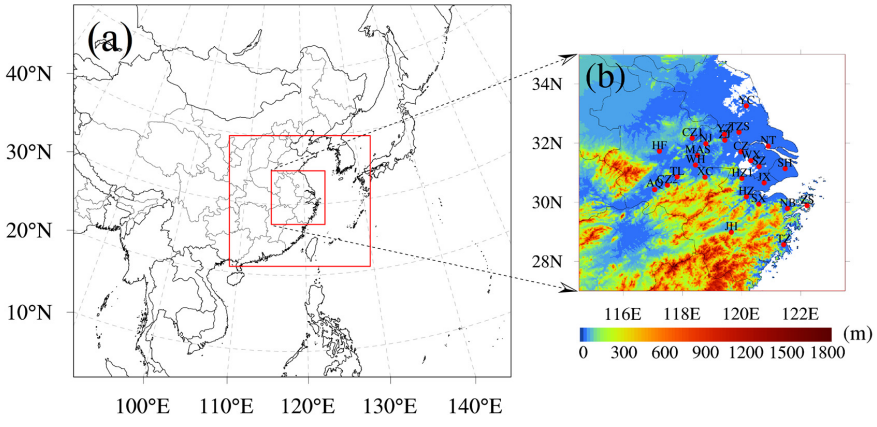

Figure 1. The three nested modeling domains (a) in WRF, and the locations of 26 typical cities in the YRD with terrain elevation data (b). The cities in (b) include: Nanjing (NJ), Wuxi (WX), Changzhou (CZ), Suzhou (SZ), Nantong (NT), Yancheng (YC), Yangzhou (YZ), Zhenjiang (ZJ) and Taizhoushi (TZS) located in Jiangsu province; Hangzhou (HZ), Ningbo (NB), Jiaxing (JX), Huzhou (HZ1), Shaoxing (SX), Jinhua (JH), Zhoushan (ZS) and Taizhou (TZ) located in Zhejiang province; Hefei (HF), Wuhu (WH), Maanshan (MAS), Tongling (TL), Anqing (AQ), Chuzhou (CZ1), Chizhou (CZ2) and Xuancheng (XC) located in Anhui province; and the

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112 **megacity Shanghai (SH). The terrain elevation data are available at**
113 **https://www.ngdc.noaa.gov/mgg/global/relief/ETOPO1/data/bedrock/cell_registered/netcdf/.**
114
115 **2 Data and methods**
116 **2.1 Air quality data**
117 Surface air pollutants monitored by the China National Environmental Monitoring Center
118 (CNMC) Network are used in this study. The nationwide observation network began operating in
119 74 major cities in 2013, and it included 1597 nonrural sites covering 454 cities by 2017 (Lu et al.,
120 2018). The monitoring data are strictly in accordance with the national monitoring regulations
121 (http://www.cnemc.cn/jcgf/dqhj/), and can be acquired from the national urban air quality real-time
122 publishing platform (http://106.37.208.233:20035/). Each monitoring site automatically measures
123 hourly air pollutants ($PM_{2.5}$, PM_{10}, SO_2, NO_2, O_3 and CO), and the urban hourly pollutants are
124 calculated by averaging the pollutants measured at all monitoring sites in that city. The MDA8 O_3
125 is calculated based on the hourly O_3 with more than 18-h measurements (Liao et al., 2017). Manual
126 inspection, including the identification and handling of invalid and lacking data, is performed
127 following previous studies (Xie et al., 2016; Shu et al., 2017; Zhan et al., 2019).
128 **2.2 Surface and sounding meteorological data**
129 With respect to surface observed meteorological data, stations at the three provincial capital
130 cities (Nanjing, Hangzhou and Hefei) and the megacity Shanghai are selected, which are ZSNJ
131 (32.00°N, 118.80°E), ZSHC (30.23°N, 120.17°E), ZSOF (31.87°N, 117.23°E) and ZSPD (31.12°N,
132 121.77°E), respectively. These surface observations, including 2-m temperature, 10-m wind speed
133 and direction and 2-m relative humidity, are recorded hourly and can be obtained from the website
134 of the University of Wyoming (http://weather.uwyo.edu/surface/).
135 To verify the upper-air fields, the sounding observations at Shanghai (31.40°N, 121.46°E) and
136 Nanjing (32.00°N, 118.80°E) are used. These sounding observations (pressure, temperature, relative
137 humidity, wind direction and wind speed etc.) are also acquired from the website of the University
138 of Wyoming (http://weather.uwyo.edu/upperair/sounding.html), with a time resolution of 12 h
139 (00:00 and 12:00 UTC).
140 **2.3 The best-track TC dataset**
141 To capture the characteristics of landfall typhoons, the best-track TC dataset issued by the

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China Meteorological Center (CMA) is considered due to its good performance on the landfall typhoons in the mainland China (available at http://tcdata.typhoon.org.cn/zjljsjj_sm.html). The dataset covers seasons from 1949 to the present, the region north of the equator and west of 180°E, and is updated annually (Li and Hong, 2016; Ying et al., 2014). A wealth of information on typhoon is recorded every 6h in the dataset, including location, minimum sea level pressure, etc. For landfall typhoons, 24h before they land and during their activities in the mainland China, the data will be recorded every 3h. Refer to the national standard for grade of tropical cyclones (GB/T 19201-2006), the intensity category (IC) of tropical cyclones is given in the dataset, which is based on the near surface maximum 2-min mean wind speed near the tropical cyclone center, ranges from 1 to 6 (Table 1).

Table 1. The intensity category of tropical cyclones

Intensity category (IC)	The near surface maximum 2-min mean wind speed near the tropical cyclone center (m/s)	Beaufort scale
Tropical depression (IC=1)	10.8-17.1	6-7
Tropical storm (IC=2)	17.2-24.4	8-9
Severe tropical storm (IC=3)	24.5-32.6	10-11
Typhoon (IC=4)	32.7-41.4	12-13
Severe typhoon (IC=5)	41.5-50.9	14-15
Super typhoon (IC=6)	≥ 51.0	≥16

2.4 Model description and configurations

To simulate the high O_3 episodes over the YRD during the period with typhoons, the WRF-CMAQ one-way coupled model is applied, which consists of WRF v3.6.1 (https://www2.mmm.ucar.edu/wrf/users/) developed by the United States National Center for Atmospheric Research (NCAR) and CMAQ v5.0.2 (https://github.com/USEPA/CMAQ/tree/5.0.2) developed by the United States Environmental Protection Agency (EPA).

WRF generates offline meteorological inputs for CMAQ with initial and boundary conditions from the National Centers for Environmental Prediction (NCEP) global final analysis fields every

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6 h at a spatial resolution of 1° × 1° (https://rda.ucar.edu/datasets/ds083.2/). Three nested domains are used, with horizontal resolutions of 81, 27 and 9 km, and grids of 88 × 75, 85 × 79 and 97 × 97, respectively (Figure 1a). There are 24 vertical sigma layers from surface to 100 hPa, with about 8 layers located below 1.5 km to resolve the boundary layer processes. Furthermore, the major physical options for the dynamic parameterization in WRF are summarized in Table 2.

Table 2. The domains and physical options for WRF in this study

Items	Contents
Dimensions (x, y)	(88, 75), (85, 79), (97, 97)
Grid spacing (km)	81, 27, 9
Microphysics	WRF Single-Moment 5-class scheme (Hong et al., 2004)
Longwave radiation	RRTM scheme (Mlawer et al., 1997)
Shortwave radiation	Goddard scheme (Kim and Wang, 2011)
Surface layer	Moni-Obukhov scheme (Monin and Obukhov, 1954)
Land-surface layer	Noah land-surface model (Chen and Dudhia, 2001)
Planetary boundary layer	YSU scheme (Hong et al., 2006)
Cumulus parameterization	Grell-Devenyi ensemble scheme (Grell and Devenyi, 2002)

Since the horizontal domains of CMAQ are one grid smaller than WRF, all three nested domains are adjusted automatically. The vertical layers of CMAQ are the same as WRF. The Meteorology Chemistry Interface Processor (MCIP) can convert WRF outputs to the necessary meteorological inputs for CMAQ. Moreover, the CB05 gas-phase mechanism with aqueous/cloud chemistry is selected in the CMAQ configurations.

The anthropogenic emissions are from the Multi-resolution Emission Inventory for China (MEIC) in 2016 with the resolution of 0.25° (http://meicmodel.org/), including anthropogenic emissions from power generation, industry, agriculture, residential and transportation sectors. All emission estimates are spatially allocated to the relevant grid cells based on the meteorological fields obtained from WRF, and are temporally distributed on an hourly basis. The simulation starts from 00:00 UTC on 13 July to 00:00 UTC 27 August, with the first 72 h as spin-up time.

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182 **2.5 Integrated process rate (IPR) analysis**

183 To quantify the contributions of individual processes in O_3 formation, the IPR analysis
184 provided in the CMAQ is utilized. The IPR analysis can illustrate the contributions to changes in
185 pollutant concentrations from seven different types of processes, including horizontal advection
186 (HADV), vertical advection (ZADV), horizontal diffusion (HDIF), vertical diffusion (VDIF), dry
187 deposition (DDEP), cloud processes with the aqueous chemistry (CLDS) and chemical reaction
188 process (CHEM), with a mass conservation adjustment at each model grid cell. The IPR analysis
189 has been widely applied to investigate regional air pollution (Fan et al., 2015; Li et al., 2012; Wang
190 et al., 2010). In this study, MADV is defined as the sum of HADV and ZADV, and TDIF is defined
191 as the sum of HDIF and VDIF.

192 **2.6 Model evaluation**

193 To evaluate the model performance, the simulation results in the innermost domain, including
194 O_3 concentration, air temperature at 2 m (T_2), relative humidity (RH), wind speed at 10 m (WS_{10})
195 and wind direction at 10 m (WD_{10}), are examined against the hourly observations at the
196 representative cities (Table 3). The statistical metrics, including correlation coefficient (R), root-
197 mean-square error (RMSE) and normalized mean bias (NMB), are used in this study. They are
198 defined as follows:

199 $$R = \frac{\sum_{i=1}^{N}(S_i-\bar{S})(O_i-\bar{O})}{\sqrt{\sum_{i=1}^{N}(S_i-\bar{S})^2}\sqrt{\sum_{i=1}^{N}(O_i-\bar{O})^2}}, \quad (3)$$

200 $$RMSE = \sqrt{\frac{\sum_{i=1}^{N}(S_i-O_i)^2}{N}}, \quad (4)$$

201 $$NMB = \frac{\sum_{i=1}^{N}(S_i-O_i)}{\sum_{i=1}^{N}O_i}\times 100\%, \quad (5)$$

202 where S_i and O_i are the simulations and observations, respectively. N is the total number of valid
203 data. $\bar{S}$ and $\bar{O}$ are the average value of simulations and observations. In general, the model results
204 are acceptable if the values of R, RMSE and NMB are close to 1, 0 and 0, respectively (Li et al.,

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2017; Shu et al., 2016; Xie et al., 2016).

2.7 Estimate of health impacts

Previous studies showed that surface O_3 pollution can induce a series of adverse health impacts by causing the incidence and mortality of respiratory diseases (Ghude et al., 2016; Jerrett et al., 2009; Lelieveld et al., 2015). To arouse more attention on the issue that O_3 can be significantly affected by typhoons in the YRD, we further estimate the premature mortality attributed to O_3 during the study period.

A standard damage function (Anenverg et al., 2010; Liu et al., 2018; Voorhees et al., 2014; WS/T 666-2019, Technical specifications for health risk assessment of ambient air pollution of China) is employed to quantify premature mortality due to O_3 exposure:

$$\Delta M = y_0(\frac{RR-1}{RR})Pop, \quad (1)$$

where ΔM is the excess mortalities attribute to O_3 exposure, y_0 is the baseline mortality rate, RR is relative risk and (RR-1)/RR is the attributable fraction, and Pop is the exposed population. RR can be calculated using the following relationship:

$$RR = \exp(\beta(C-C_0)), \quad (2)$$

where β is the concentration-response factor, C is the exposure concentration and C_0 represents the theoretical minimum-risk concentration.

In this study, the mortality rate for respiratory disease is obtained from China Health and Family Planning Statistical Yearbook 2018 (https://www.yearbookchina.com/navibooklist-n3018112802-1.html), which is 68.02/100000. The β is generated from Dong et al. (2016), that is 0.461%. The population data are obtained from the bureau of statistics of different cities in the YRD. The C_0 is 70 $\mu g\ m^{-3}$ for MDA8 O_3 given by the World Health Organization (WHO).

3 Results and discussions

3.1 Characteristic of O_3 episodes

In the midsummer season, the warm sea surface (high temperature) is conducive to the generation of typhoon (high O_3 concentration), which provides a good opportunity to investigate the mechanism of typhoon affecting O_3 in the YRD. Figure 2 shows the MDA8 O_3 in the typical 26 cities of the YRD in summer of 2018. O_3 concentration tended to be high in June, and relatively low

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in July and August. The relatively low O_3 may be attributed to the maritime air masses transported by the Asian summer monsoon (Ding et al., 2008; Xu et al., 2008). Nevertheless, we notice that there are two regional multiday O_3 pollution episodes from 24 July to 11 August in the YRD, which means that about half of the cities in the YRD exceed the national air quality standard (The national ambient air quality standard for MDA8 O_3 is 160 μg m^{-3} in China). The first multiday O_3 episodes appeared in most cities from 24 July to 2 August. The highest MDA8 O_3 concentration reached up to 264 μg m^{-3} on 27 July in Ningbo (NB). O_3 pollution was even observed for 6 consecutive days from 27 July to 1 August in Maanshan (MAS). Only two days later, regional O_3 pollution occurred in the YRD again from 5 August to 11 August.

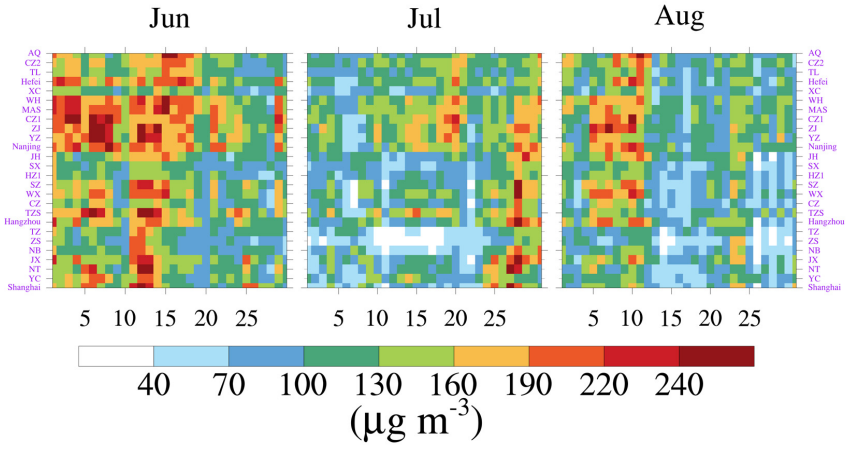

Figure 2. The MDA8 O_3 in 26 cities of the YRD in summer of 2018. The national ambient air quality standard for MDA8 O_3 is 160 μg m^{-3} in China. These cities are sorted by longitude.

Figure 3 further shows diurnal variation of O_3 in all 26 cities of the YRD from 00:00 16 July to 00:00 25 August (throughout this paper the time refers to UTC, unless LST is specifically mentioned). Interestingly, O_3 pollution occurred earlier in cities near the coastline (large longitude, Figure 1b) rather than concurrently during the two multiday O_3 episodes. For example, from 24 July to 2 August, the first day that hourly O_3 concentration exceeded the national air quality standard (The national ambient air quality standard for hourly O_3 is 200 μg m^{-3} in China) in Shanghai, Hangzhou, Nanjing and Hefei was 24 July, 27 July, 28 July and 31 July, respectively. Thus, we

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classify the 26 cities in the YRD into four categories based on their longitudes, surrounding the four representative cities (Figure 4). The category I cities include SH, YC, NT, JX, NB, ZS and TZ. The category II cities include HZ, TZS, CZ, WX, SZ, HZ1, SX and JH, and the category III cities include NJ, YZ, ZJ, CZ1, MAS, WH and XC. Other cities are classified as the category IV cities, which are HF, TL, CZ2 and AQ. The first category cities are closest to the coastline, while the fourth category is the opposite.

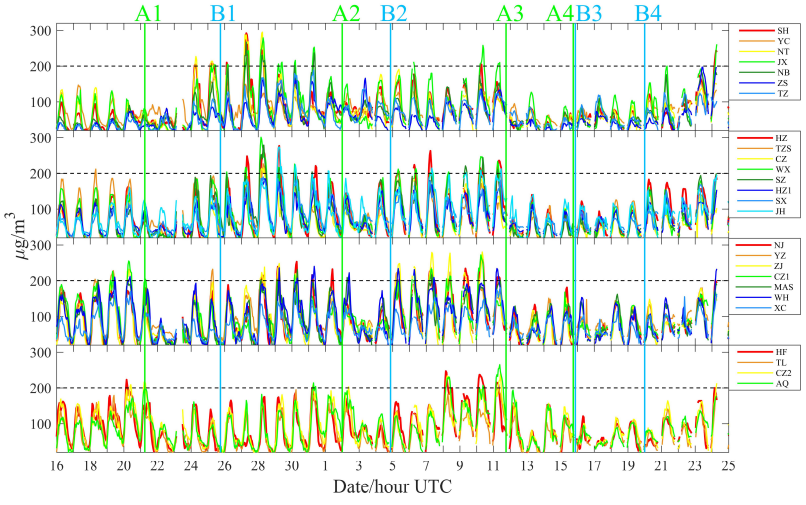

Figure 3. Diurnal variation of O_3 in all 26 cities of the YRD. The grey dotted lines are the national ambient air quality standard for hourly O_3 (200 μg m^{-3}) in China. The letter A indicates the moment that the typhoon has reached the 24-h warning line, and letter B indicates the last moment of typhoon activity in the mainland China. These moments are acquired from the best-track TC dataset, depending on the start and end time of the densified observations. Coordinates 1, 2, 3, and 4 represent Typhoon Ampil, Typhoon Jongdari, Typhoon Yagi, and Typhoon Rumbia, respectively. Note: these cities are sorted by longitude.

3.2 Landfall typhoons and their effects

For such O_3 episodes with regional, long-lasting characteristics, may often be associated with slow-moving synoptic weather systems. We find that the O_3 episodes coincided well with landfall typhoons activities, and the track and intensity of typhoons are given in Figure 4. Typhoon Ampil

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was first observed at 00:00 on 18 July, and landed in Shanghai around 4:30 on 22 July with an intensity of severe tropical storm (IC=3). During the time of Typhoon Ampil, Typhoon Jongdari generated over the western North Pacific at 12:00 on 23 July, and made landfall at the junction of Zhejiang province and Shanghai at 21:00 1 August. After Typhoon Jongdari, Typhoon Yagi generated at 00:00 7 August. At around 15:35 12 August, it landed in Zhejiang province and remained active in the mainland China until 21:00 15 August. Before the end of Typhoon Yagi, Typhoon Rumbia was observed over the western North Pacific at 6:00 14 August. It finally landed in Shanghai at around 20:00 16 August, causing huge economic losses.

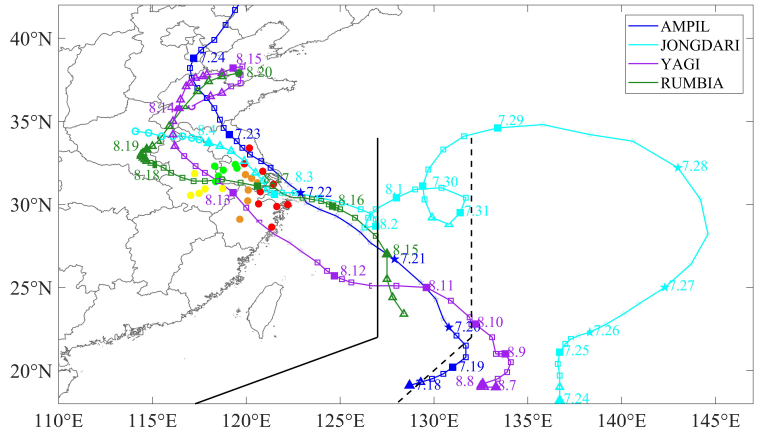

Figure 4. The track and intensity of Typhoon Ampil, Typhoon Jongdari, Typhoon Yagi, and Typhoon Rumbia. The circle, triangle, square and pentagram indicate the intensity category of tropical cyclones is less than 1 (IC < 1), equal to 1 (IC = 1), equal to 2 (IC = 2), and not less than 3 (IC >= 3), respectively. Black solid line and dotted line represent the 24-hour and 48-h warning line for tropical cyclones, respectively. The colored solid points are the locations of cities in the YRD, and different color represents different cities categories. Wherein, red, carrot, green and yellow are category I, II, III and IV cities, respectively.

To further understand the relationship between O_3 episodes and landfall typhoons, we mark the critical moments of landfall typhoons in Figure 3. The letter A indicates the moment that typhoons have reached the 24-h warning line, and the letter B indicates the last moment of typhoon activity

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296 in the mainland China. These moments are acquired from the best-track TC dataset, depending on
297 the start and the end time of the densified observations. Coordinates 1, 2, 3, and 4 represent Typhoon
298 Ampil, Typhoon Jongdari, Typhoon Yagi, and Typhoon Rumbia, respectively. As shown in Figure
299 3, O_3 exhibited a significant cycle during the study period. That is, when the typhoon is close enough
300 (near moments A1, A2, A3 and A4), the O_3 concentrations decreased, but O_3 concentrations would
301 increase as long as the typhoon was not active in the mainland China (B1, B2 and B4) any more.
302 This cycle would repeat if the next typhoon approached. O_3 pollution was likely to occur during the
303 period from the end of the typhoon to the arrival of the next typhoon (B1A2 and B2A3) in the YRD.
304 Furthermore, we find that the variations of O_3 was related to the track, duration and landing
305 intensity of the typhoons. For example, during the B1A2 period when the O_3 pollution occurred, the
306 moments that hourly O_3 concentrations first exceed 200 μg m^{-3} in about half of cities of the
307 categories I, II, III and IV were 6:00 UTC (14:00 LST) 27 July, 6:00 UTC (14:00 LST) 28 July, 3:00
308 UTC (11:00 LST) 29 July and 6:00 UTC (14:00 LST) 31 July, respectively. This phenomenon also
309 suggests that O_3 pollution occurs in coastal region will be ahead of that in inland regions, which
310 may be related to the track of typhoons (Figure 4). As for the impact of typhoon duration, the A4B3
311 period provided a good interpretation. While Typhoon Yagi was still active in the mainland China,
312 Typhoon Rumbia had reached the 24-hour warning line. Hence, the O_3 remained a low level
313 throughout the period (A3B4), which was quite different from B1A2 and B2A3 period. Noted that
314 the landing point and active path of Typhoon Ampil and Typhoon Jongdari were very similar (Figure
315 4). However, the landing intensity of Typhoon Ampil was severe tropical storm (IC = 3), and that of
316 Typhoon Jongdari was tropical storm (IC = 2), resulting in a difference in O_3 concentrations for
317 Shanghai. Within 24 hours after Typhoon Ampil (Jongdari) reached the 24-hour warning line, the
318 average O_3 concentrations was 40.9 (80.1) μg m^{-3} in Shanghai. This is because that the stronger the
319 typhoon landed, the gale (The 10-m wind speed near moment A1 was larger than that near moment
320 A2 in Shanghai, Figure 7a) and precipitation accompanying the typhoon will be more effective in
321 removing O_3.

322 **3.3 Processes of O_3 pollution affecting by typhoons**

323 To reveal the major processes of O_3 pollution episodes affected by landfall typhoons, four
324 representative cities (Shanghai, Hangzhou, Nanjing and Hefei) are selected for further analysis –
325 based on monitoring data and model results.

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326 **3.3.1 Evaluation of model performance**

327 To evaluate the simulation performance, the hourly simulation results are compared with the
328 measurements during 00:00 16 July to 00:00 25 August. Table 3 presents the statistical metrics for
329 selected variables, including temperature at 2 m (T_2), relative humidity (RH), wind speed at 10 m
330 (WS_{10}) and wind direction at 10 m (WD_{10}), and surface O_3. T_2 is reasonably well simulated, with R
331 values of 0.75, 0.77, 0.72 and 0.64 in Shanghai, Hangzhou, Nanjing and Hefei, respectively. Though
332 our simulation underestimates T_2 to some certain extent, the slightly underestimation is acceptable
333 due to the small RMSE (3.2, 2.7, 2.9 and 3.3) and NMB (-7.5%, -5.1%, -5.5% and -5.5%) values.
334 As for RH, the simulation results are overestimated in all four cities, leading to the NMB values of
335 9.1%, 4.6%, 6.7% and 0.5% in Shanghai, Hangzhou, Nanjing and Hefei, respectively. With high R
336 values (0.69, 0.65, 0.71 and 0.71) and relatively low RMSE values (12.4, 12.8, 12.1 and 10.8), the
337 WRF simulates RH over the YRD quite well. The wind fields are closely related to the transport
338 processes of air pollutants. The overestimation of WS_{10} may partly be attributed to the unresolved
339 terrain features by the default surface drag parameterization causing overestimation of wind speed
340 in particular at low values (Jimenez and Dudhia, 2012; Li et al., 2017). With regards to WD_{10}, the
341 simulation error is large based only on these statistical metrics. This is because that near-surface
342 wind fields are deeply influenced by local underlying surface characteristics, and improving the
343 urban canopy parameters might be useful (Liao et al., 2015; Xie et al., 2016). In term of O_3, the
344 simulation results for O_3 concentrations behave satisfactorily. R is as high as 0.55, 0.65, 0.66 and
345 0.54 in Shanghai, Hangzhou, Nanjing and Hefei, respectively, while the NMB values are 5.8%,
346 16.4%, -6.2% and -5.3%, respectively.

347

348 **Table 3. Statistical metrics for meteorological and chemical variables.**

City	Variable	$\bar{O}$	$\bar{S}$	R	RMSE	NMB
Shanghai	T_2 (℃)	30.3	28.1	0.75	3.2	-7.5%
	RH (%)	75.0	81.8	0.69	12.4	9.1%
	WS_{10} (m s^{-1})	4.9	5.5	0.51	2.3	11.7%
	WD_{10} (°)	144.8	113.4	0.01	113.5	-22.9%
	O_3 (μg m^{-3})	74.3	76.5	0.55	45.3	5.8%
Hangzhou	T_2 (℃)	30.3	28.8	0.77	2.7	-5.1%
	RH (%)	75.1	78.5	0.65	12.8	4.6%
	WS_{10} (m s^{-1})	3.3	4.7	0.32	2.7	32.5%

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	WD_{10} (°)	155.0	114.7	-0.10	132.5	-27.8%
	O_3 (μg m^{-3})	81.7	91.3	0.65	49.8	16.4%
Nanjing	T_2 (℃)	29.8	28.1	0.72	2.9	-5.5%
	RH (%)	77.4	82.6	0.71	12.1	6.7%
	WS_{10} (m s^{-1})	3.1	5.0	0.39	3.0	63.8%
	WD_{10} (°)	132.8	115.6	0.21	102.7	-15.0%
	O_3 (μg m^{-3})	87.6	79.8	0.66	46.7	-6.2%
Hefei	T_2 (℃)	29.3	27.7	0.64	3.3	-5.5%
	RH (%)	81.1	81.5	0.71	10.8	0.5%
	WS_{10} (m s^{-1})	3.2	3.2	0.37	2.2	2.9%
	WD_{10} (°)	147.0	128.6	0.04	136.7	-13.3%
	O_3 (μg m^{-3})	87.3	80.3	0.54	45.0	-5.3%

Note. R exceeds 0.1 to reach statistically significant at 99.9% confident level.

Figure 5 further shows hourly variations of O_3, T_2, WS_{10} and WD_{10} for measurements and simulations in four representative cities. The simulations effectively reproduce the diurnal variation of O_3, T_2 and WS_{10}, confirming the reliability of the simulation results. Moreover, the model well captures the shift in wind direction during the study period. Thus, the overall model performance for wind fields is acceptable. In summary, the simulation results can capture and reproduce the major characteristics of the O_3 episodes, including the meteorological conditions and evolution of O_3, which can provide valuable insights into the formation of the O_3 episodes.

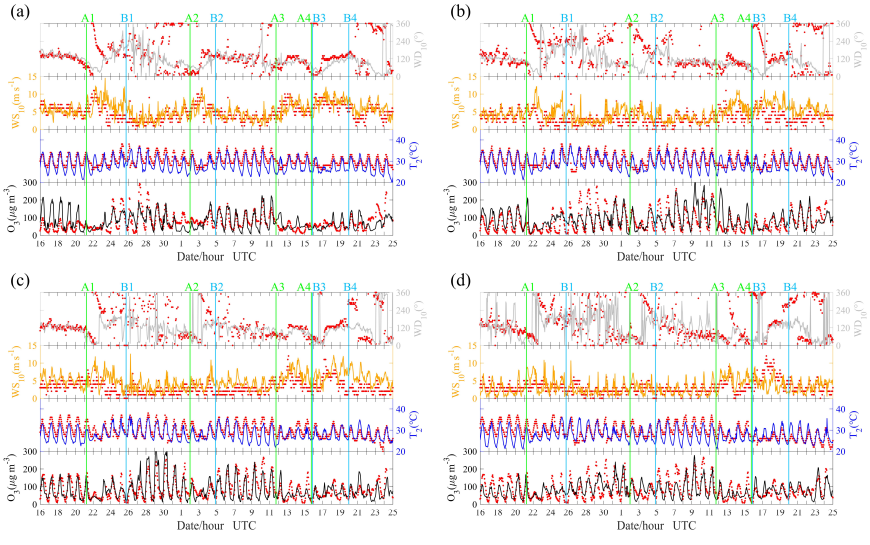

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Figure 5. Hourly variations of O_3, T_2, WS_{10} and WD_{10} for measurements (red dots) and simulation (colored lines) in (a) Shanghai, (b) Hangzhou, (c) Nanjing and (d) Hefei.

3.3.2 Shanghai in category I cities

In this study, Shanghai was usually one of the first cities affected by landfall typhoons. We can see a multiday episode of O_3 during the period of 24-28 July, with a maximum of hourly O_3 up to 292 μg m^{-3} at 27 July (Figure 6a). The high O_3 concentrations together with high primary pollutants (CO and NO_2) suggest a strong photochemical O_3 production under the condition of high temperature (The daily maximum temperature can reach 35 ℃) during this period, and the weak wind may play a significant role in the accumulation of surface O_3. The increase of primary pollutants may be related to the wind shift from southeast to southwest causing by Typhoon Ampil (A1 in Figure 6a, -A1 and A1B1 in Figure 7), resulting in air masses originally from the ocean had become inland. Interestingly, $PM_{2.5}$ also showed good correlation with O_3 and primary pollutants, especially for NO_2 during this period. This indicates that a high level of oxidizability can promote the formation of secondary particles (Kamens et al., 1999; Khoder, 2002). From the results of process analysis (Figure 6b), the major contributions to surface O_3 were TDIF, CHEM and DDEP due to the small net contribution of MADV. TDIF had a considerable positive contribution while DDEP did the opposite, suggesting that high surface O_3 may be sourced from the upper layer via TDIF process, and be removed via DDEP process. However, for the whole boundary layer, which is defined as the layer less than 1500 m in this study, it was the balance between CHEM and DDEP instead TDIF and DDEP. Thus, TDIF was likely to play the role of "transport" from the upper layer to surface. Figure 6c further shows the temporal-vertical distribution of O_3 with vertical wind velocity. The downward airflows were prevailed over Shanghai until 23 July, which are induced by the subtropical high. Then, strong upward airflows appeared as Typhoon Ampil arrived, and high level of O_3 disappeared. Around 27 July, the downward airflows gradually resumed and high level of O_3 occurred. It is note-worthy that the high value center of O_3 appeared near the altitude of 1 km instead of near surface, indicting high photochemical production efficiency of O_3 occurred in the middle boundary layer. The downward airflows can not only inhibit the vertical transport of O_3 but also transport high-level O_3 to the surface, causing the episodes of surface O_3.

As shown in Figure 7, O_3 pollution tends to occur during the period from the end of the typhoon

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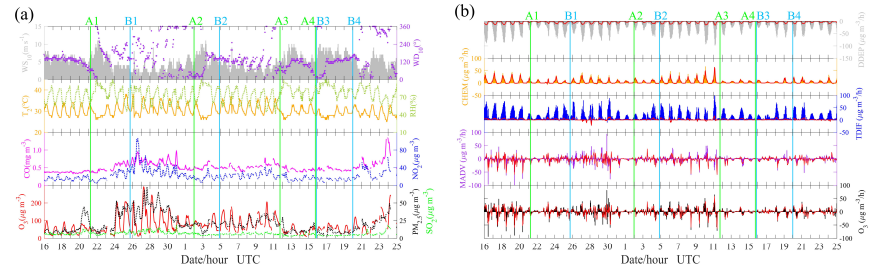
391 to the arrival of the next typhoon (B1A2 and B2A3) in the YRD. To reveal this phenomenon, we
392 compare these two periods (B1A2 and B2A3) with their previous periods (A1B1 and A2B2) using
393 the skew-T log-P diagram (Figure 6d and 6e). It is found that the atmospheric conditions of B1A2
394 (B2A3) were hotter and drier than A1B1 (A2B2) below 700 hPa in Shanghai, and wind speed is
395 smaller in B1A2 (B2A3). Those changes in atmospheric conditions after typhoon will be conducive
396 to the generation of high O_3 concentration in Shanghai.
397



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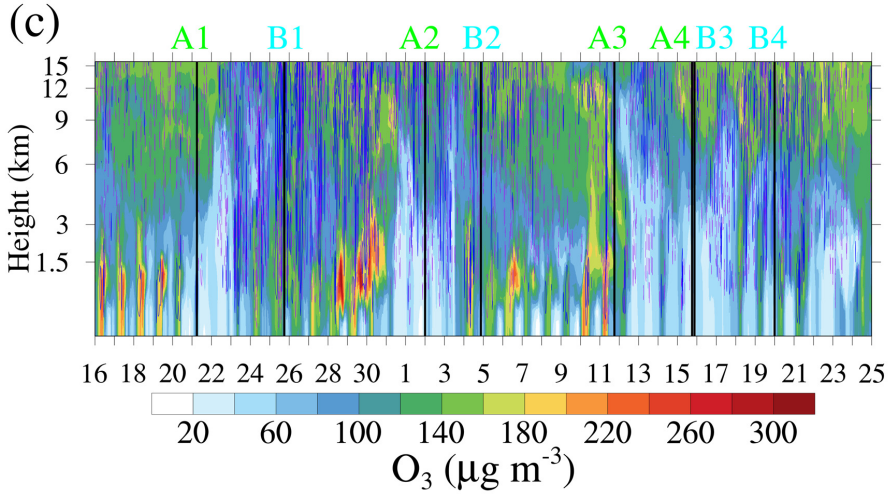

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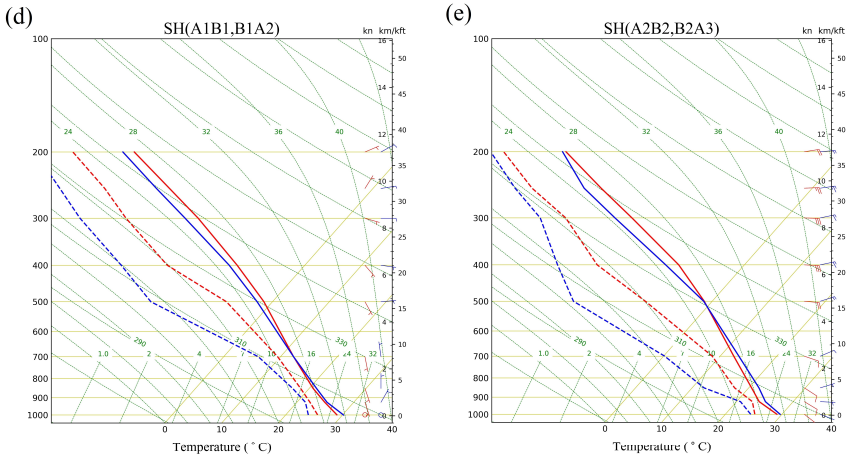

Figure 6. (a) Time series of air pollutants (O_3, $PM_{2.5}$, SO_2, CO and NO_2) and meteorological factors (T_2, RH, WS_{10} and WD_{10}) in Shanghai. (b) Individual processes contribution to net O_3 density at Shanghai. O_3 is the net increase, MADV is the sum of horizontal advection (HADV) and vertical advection (ZADV), TDIF is the sum of horizontal diffusion (HDIF) and vertical diffusion (VDIF), CHEM is chemical reaction process, and DDEP is dry deposition process. The color histograms indicate the results for the layer near the surface, while the solid red lines indicate the average results for all layers below 1500 m. (c) Temporal-vertical distribution of O_3 with vertical wind velocity over Shanghai. The dotted purple line and solid blue line indicate the negative wind speeds (downward airflows) and positive wind speeds (upward airflows), respectively. (d) The skew-T log-P diagram at Shanghai. The average results of period A1B1 and B1A2 are shown in red and blue, respectively. (e) Same as (d), but for the average results of period A2B2 and B2A3.

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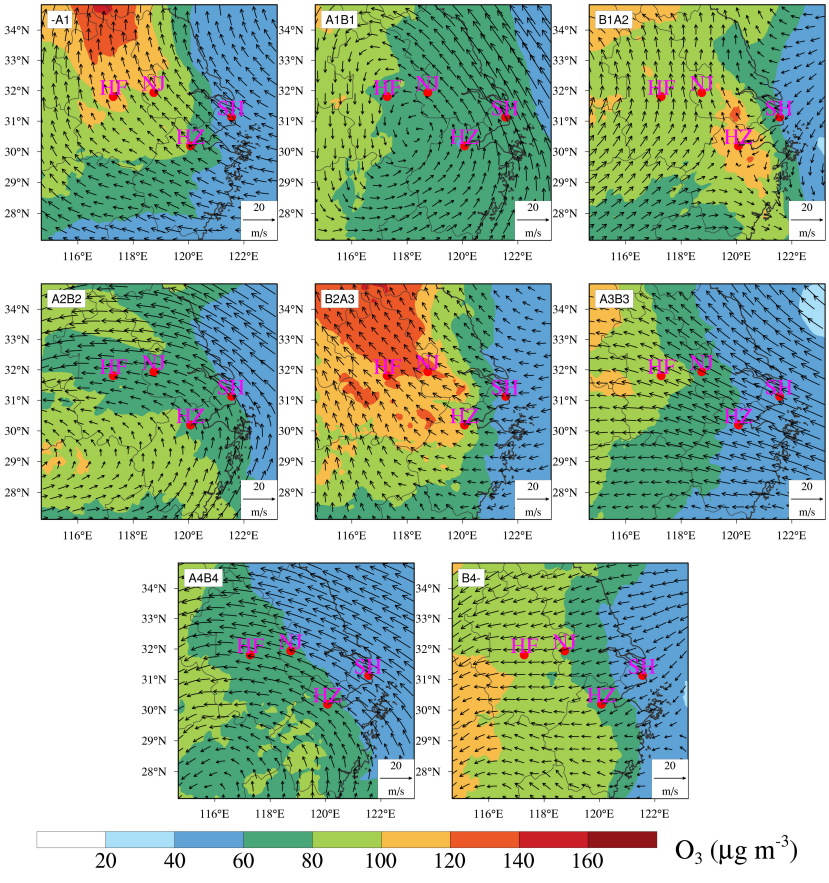

Figure 7. Spatial distribution of surface O_3 with wind fields at 850 hPa over the YRD. -A1, A1B1, B1A2, A2B2, B2A3, A3B3, A4B4, B4- are the average results from the beginning to A1, A1 to B1, B1 to A2, A2 to B2, B2 to A3, A3 to B3, A4 to B4, B4 to the end, respectively. Details can be found in Figure 4.

3.3.3 Hangzhou in category II cities

Figure 8 presents the results for Hangzhou. It shows that high O_3 concentrations occurred on 27-31 July and 5-7 August, which may also be related to the strong photochemical production of O_3 under the abundance of precursors (Figure 8a) and poor diffusion conditions due to the light wind (B1A2 and B2A3 in Figure 7). Figure 8a further shows that high O_3 was often associated with an increase in CO but the NO_2 concentrations usually remained at the same level. This phenomenon indicates a VOCs-limited regime in this city since CO usually have good correlation with VOCs

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and can play a similar role as VOCs in the photochemical production of O_3 (Atkinson, 2000; Ding et al., 2013). In fact, O_3 in other representative cities (Shanghai, Nanjing and Hefei) also showed a better correlation with CO than NO_2. Though Hangzhou is close to Shanghai, there is a significant difference of wind fields over these two cities. Starting from the arrival of Typhoon Ampil (A1). The wind direction in Hangzhou did not change back to southeast until a few days later after Typhoon Jongdari dissipated (B2). During this period (A1B2), the frequent southwest wind may be the reason for high CO concentrations in Hangzhou. In addition, the chaotic wind field during period B1A2 (B1A2 in Figure 7) may lead to the light wind in Hangzhou. With respect to the simulation results, the model simulated the variation of O_3 but failed to capture the O_3 peaks (e.g., the peak values on 27-31 July), which may be related to the strong upward airflows (Figure 8c) that inhibited the accumulation of O_3 (Figure 8b). This further illustrates that downward airflows may be an important factor for O_3 episodes in this case.

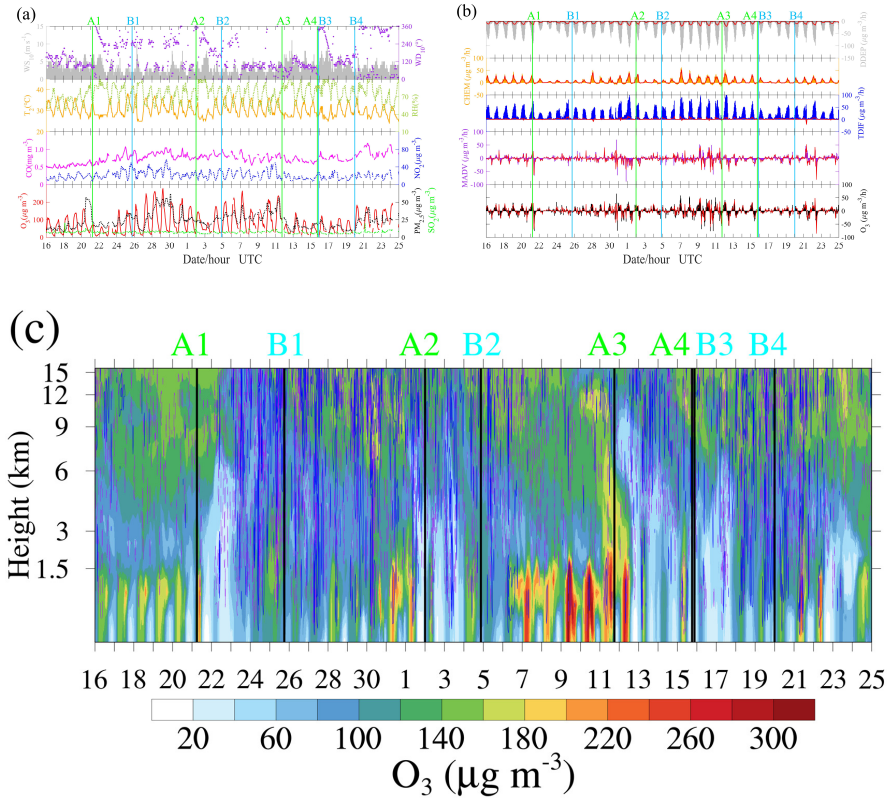

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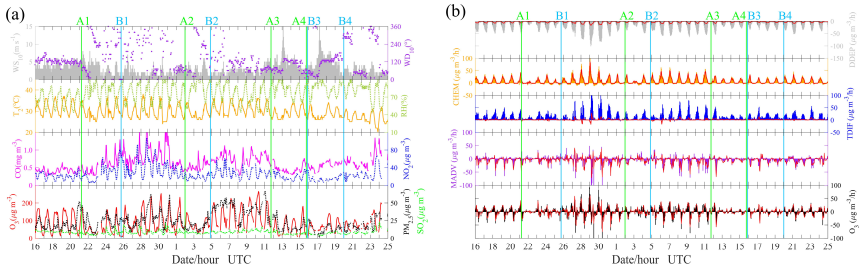
Figure 8. Same as Figure 6 (a)-(c), but for Hangzhou.

3.3.4 Nanjing in category III cities

As for Nanjing, the O_3 episodes with exceedance of national air quality standards were observed on 28 July to 1 August and 7-11 August. These O_3 episodes were characterized by abundant O_3 precursors under the condition of high temperature. Furthermore, light wind (B1A2 and B2A3 in Figure 7) and downward airflows (Figure 9c) also contributed greatly to the occurrence of O_3 pollution, with the similar mechanism as that of Shanghai and Hangzhou. As early as 22 July, the wind direction in Nanjing had changed from southeast to southwest affected by Typhoon Ampil, and the concentrations of the main primary pollutants (CO, NO_2 and SO_2) increased (Figure 9a). However, high-level O_3 episodes did not occur until 28 July even though the maximum temperature did not change significantly during 24-31 July. The "obstacle" of the O_3 episodes may be the precipitation causing by the strong upward airflows during 23-26 July (Figure 9c). As shown in Figure 9b, high surface O_3 concentration during the pollution episodes is the result of TDIF and CHEM processes, and is lost through DDEP and MADV processes. With respect to the vertical structure of atmospheric, B1A2 (B2A3) was also hotter and drier than A1B1 (A2B2) below 700 hPa in Nanjing (Figure 9d and 9e). The similar results as Shanghai further confirm that high O_3 concentrations are more likely to occur during the period from the end of the typhoon to the arrival of the next typhoon (B1A2 and B2A3) than the period when the typhoon approaches and is active in the mainland China (A1B1 and A2B2).



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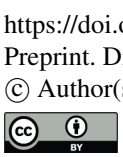

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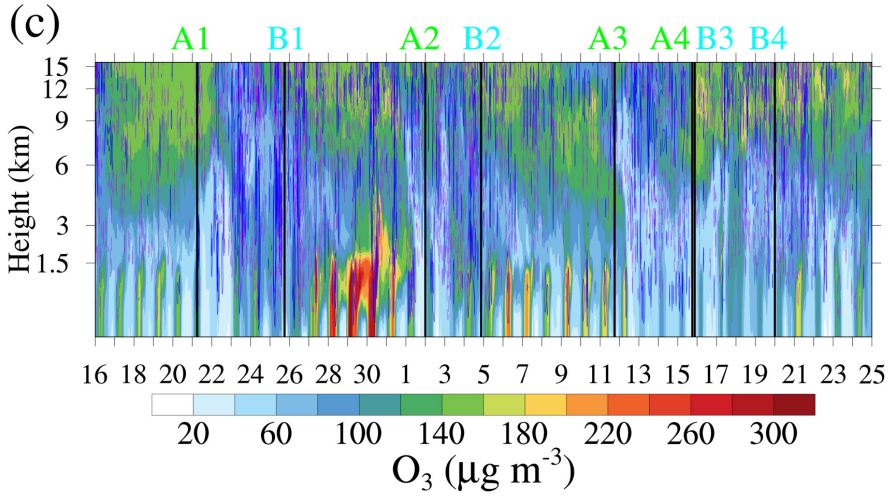

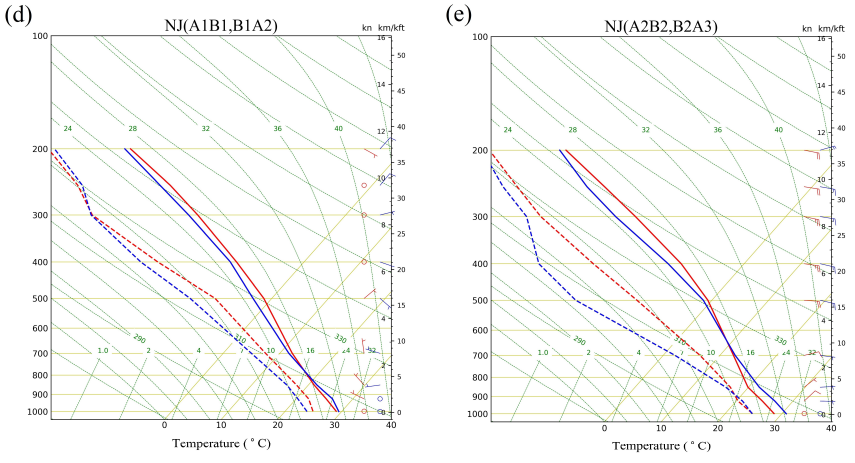

Figure 9. Same as Figure 6, but for Nanjing.

3.3.5 Hefei in category IV cities

Hefei is the city farthest from the coast among four representative cities, and O_3 pollution occurred on 31 July and 8-11 August. We can also find the phenomenon that the precursors concentrations had an increase once the wind direction changed from southeast to southwest (Figure 10a). During B1A2 and B2A3, the main precursors of O_3 had a high level. However, high O_3 concentration was mainly found in B2A3, and not in B1A2. This may be related to the relatively low temperature during B1A2 (Figure 10a), which is not conducive to photochemical production of

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O_3 (Figure 10b). As shown in Figure 10c, there were distinct upward airflows within the boundary layer, which may be related to urban effect (e.g., urban heat islands). These upward airflows within the boundary layer help mix the air, resulting in a uniform distribution of O_3 in the vertical direction. However, the downward airflows can still inhibit the vertical diffusion of O_3, and O_3 tends to be trapped within the boundary layer.

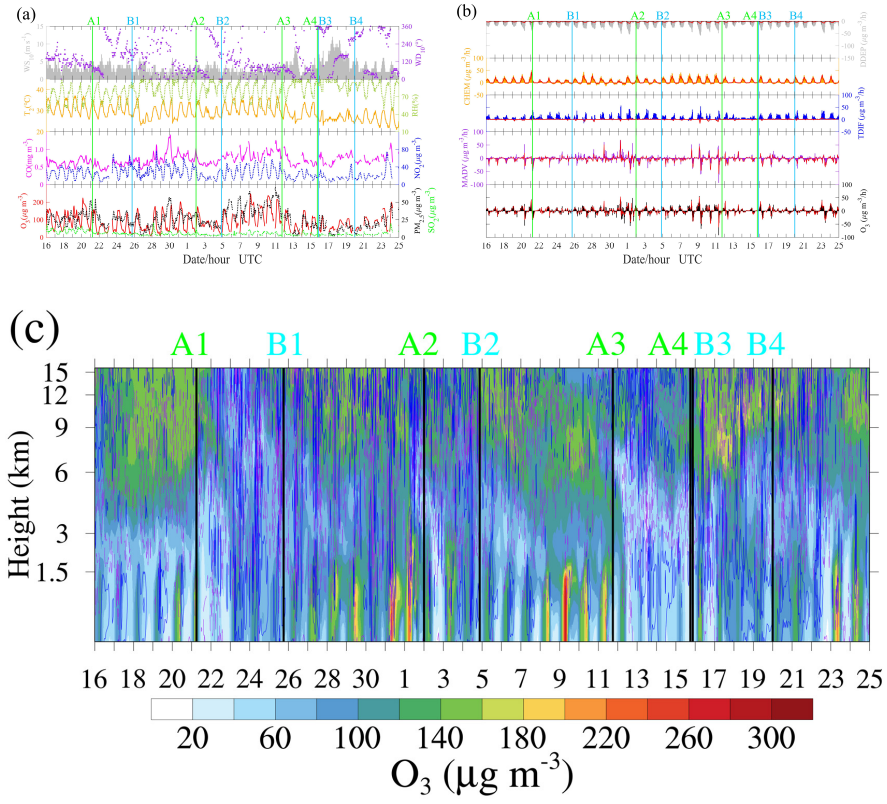

Figure 10. Same as Figure 6 (a)-(c), but for Hefei.

3.3.6 A schematic diagram of major processes

Although the processes of landfall typhoon affecting O_3 varied from city to city, the major processes have many similarities and can be summarized as a schematic diagram in Figure 11. The YRD region, which features a typical subtropical monsoon climate, is strongly influenced by the western Pacific subtropical high in summer. Dominated by the subtropical high, the meteorological

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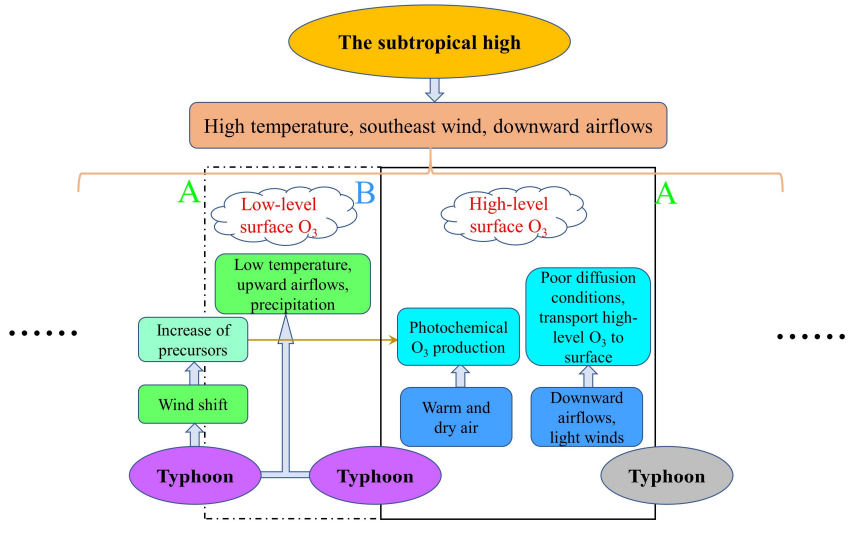
conditions of high temperature and downward airflows combined with high levels of precursors due to the huge energy consumption tend to form high O_3 concentrations in this region. However, powerful systems like typhoon can break this state. For typhoons that may land in the YRD, by the time they approach the 24-hour warning line, the prevailing southeast wind in the YRD will change to southwest wind, which can transport lots of precursors from inland to the YRD. The change in wind direction depends on the track of typhoon and the geographical location of cities, and often appears first in coastal region. With typhoon, the low temperature, precipitation (upward airflows) and wild wind will prevent high O_3 and $PM_{2.5}$ episodes. Moreover, the effect of removing pollutants is related to the intensity of typhoon landing, but some of the main precursors of O_3 are still at a high level due to foreign sources superposed with local emissions. After typhoon, the atmosphere returns to a warm and dry state (even more so than before), and strong photochemical reactions begin to produce O_3 under the abundance of precursors. O_3 is mainly generated inside the boundary layer (~1000 m), and then transported to the surface by downward airflows or turbulent mixing. At the same time, the wind readjusts to southeast and wind speed is light, resulting in poor diffusion conditions. The downward airflows and light wind obstruct the vertical and horizontal diffusion of O_3, leaving O_3 trapped on the ground. The thermal-dynamic effects result in high-level surface O_3 in the YRD.



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509 **Figure 11. A schematic diagram of major processes that summertime O_3 affected by landfall**
510 **typhoon in the YRD. The letter A indicates the moment that the typhoon has reached the 24-**
511 **h warning line, and letter B indicates the last moment of typhoon activity in the mainland**
512 **China.**
513
514 In fact, most typhoons generated over the western North Pacific will not land in China, or they
515 are more likely to land in the South China rather than the YRD. In our previous study (Shu et al.,
516 2016), the typhoon did not land in the YRD, but the processes of high-level O_3 formation may be
517 common. That is, the processes in the dashed box in Figure 11 are unique to landfall typhoons, while
518 the processes in the solid box can be found as long as the typhoons that can affect the YRD.
519 Transport of precursors, downward airflows, high temperature and light wind are crucial factors,
520 and the roles of those factors play in O_3 episodes depends on typhoon and city. It is hard to quantify
521 these processes with just a few cases. For example, we cannot estimate whether the downward
522 airflows are dominated by the subtropical high or the periphery circulation of typhoons since they
523 usually occur simultaneously. Furthermore, the behave of particulate matter is intriguing since high-
524 level $PM_{2.5}$ often occurs with high-level O_3 after typhoon, which is different from previous studies
525 that high particulate matter concentrations inhibit the formation of O_3 (Li et al., 2005; Xing et al.,
526 2017). This may be related to the heterogeneous reactions (Lou et al., 2014) but research on this
527 issue is quite limited to date.
528
529 **3.3 Premature mortalities induced by O_3 exposure**
530 When it comes to typhoons, especially landfall typhoons, the first concern is the huge damage
531 caused by extreme weathers. After typhoons, people are relieved and busy with their work as usual.
532 However, our research indicates that high O_3 episodes are likely to occur in the short period after a
533 typhoon in the YRD, and high O_3 concentrations can do harm to people's health. To arouse attention
534 on this issue, we estimate the premature mortality attributed to O_3 for respiratory disease, we choose
535 two complete cycles, which is the period A1A3 (21 July to 11 August), to do the calculation. Table
536 4 summarized the premature mortalities in cities in the YRD. The premature mortalities are a
537 function of both the population and O_3 levels, resulting in high premature mortalities in populated
538 and heavily polluted areas. Out of the 26 cities in the YRD, Shanghai shows highest premature

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539 mortalities (29.2) due to its high surface O_3 concentrations and huge population. The city with the
540 lowest premature mortalities (0.6) is Zhoushan, which may be related to removing effect of the
541 maritime air masses as Zhoushan is located by the sea (Figure 1b). During this period, the total
542 premature mortalities in the YRD is 194.0, which is larger than the number of casualties caused
543 directly by the typhoons (80 people were killed by landfall typhoons in mainland China in 2018).
544

545 **Table 4. Premature mortalities induced by O_3 exposure for respiratory disease**

	City	Population (thousand)	Premature mortalities
Category I cities	Shanghai	24,240	29.2
	Yancheng	7,200	6.1
	Nantong	7,310	7.9
	Jiaxing	4,726	7.3
	Ningbo	8,202	8.1
	Zhoushan	1,173	0.6
	Taizhou	6,139	4.1
Category II cities	Hangzhou	9,806	16.5
	Taizhoushi	4,636	5.2
	Changzhou	4,729	4.4
	Wuxi	6,575	10.7
	Suzhou	10,722	15.3
	Huzhou	3,027	2.8
	Shaoxing	5,035	4.7
	Jinhua	5,604	8.2
Category III cities	Nanjing	8,436	13.4
	Yangzhou	4,531	5.5
	Zhenjiang	3,196	5.3
	Chuzhou	4,114	5.8
	Maanshan	2,337	3.6
	Wuhu	3,748	6.2
	Xuancheng	2,648	2.0
Category IV cities	Hefei	8,087	10.9
	Tongling	1,629	1.7
	Chizhou	1,475	2.1
	Anqing	4,691	6.4
Total		154,016	194.0

546

547 **4 Conclusions**

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548 In this study, we investigate the detail processes of landfall typhoons affecting O_3 in the YRD
549 based on a unique case during 16 July to 25 August with the help of monitoring observations and
550 numerical simulations. This case was characterized by two multiday reginal O_3 pollution episodes
551 concerned with four successive landfall typhoons. The two O_3 episodes appeared from 24 July to 2
552 August and 5 to 11 August, respectively, with the highest MDA8 O_3 reached up 264 μg m^{-3}.
553 The moment that typhoon reaches the 24-h warning line and the last moment of typhoon
554 activity in the mainland China are crucial, because O_3 pollution episodes mainly occurred during
555 the period from the end of typhoon and the arrival of the next typhoon in the YRD. These two
556 moments can be roughly regarded as time nodes. Furthermore, it is found that the variations of O_3
557 was related to the track, duration and landing intensity of the typhoons during the study period. O_3
558 pollution appeared in coastal region was ahead of that in inland regions due to the track of typhoons.
559 The interval between two typhoons can affect the duration of high O_3 concentration in the YRD.
560 Generally, sustained high O_3 concentration tends to appear on days when the typhoon has dissipated
561 but not influenced by the new one. As for the landing intensity of typhoon, the stronger the typhoon
562 landed, the gale and precipitation accompanying the typhoon will be more effective in removing O_3,
563 resulting in lower O_3 concentration in the typhoon landing location.
564 The detail processes of landfall typhoons affecting O_3 depend on typhoons and cities. High
565 temperature and downward airflows dominated by the subtropical high combined with abundant
566 precursors are the main reason for high O_3 concentration in the YRD in summer. And landfall
567 typhoon can change this state through the following mechanism: When the landfall typhoon is close
568 enough (~ 24-hour warning line), the prevailing southeast wind will change to southwest wind,
569 which transports large amount of precursors from inland to the YRD. The southwest wind usually
570 appears first in coastal region, and will turn back to southeast wind as long as the YRD is dominated
571 by subtropical high. Then the typhoon makes landfall, the low temperature, precipitation (upward
572 airflows) and wild wind suppress the generation of O_3. After typhoon, the atmosphere at low layer
573 (below 700 hPa) will be warm and dry, and strong photochemical reactions begin to produce O_3
574 under the abundance of precursors due to foreign sources superposed with local emissions. O_3 is
575 mainly generated in the middle of boundary layer (~ 1000 m), and then transported to the surface
576 by downward airflows or turbulent mixing. The downward airflows also obstruct the vertical
577 diffusion of O_3. Meanwhile, wind speed is light when the wind readjusts to southeast, which further

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worsens horizontal diffusion of O_3. The O_3 can be accumulated and trapped on the ground. The thermal-dynamic effects results in high surface O_3 concentration in the YRD. Those processes will repeat if the next typhoon approach.

The estimated premature mortalities attributed to O_3 exposure for respiratory disease in the YRD during 21 July to 11 August (two complete cycles of typhoons) is 194.0, which is larger than the number of casualties caused directly by the typhoons. This work can enhance our understanding of how landfall typhoons affect O_3 in the YRD, which may help to forecast the O_3 pollution synthetically impacted by the subtropical high and typhoon. Meanwhile, our results further confirm that large-scale synoptic weather systems play an important role in regional air pollution, suggesting a need in establishing potential links between air pollution and predominant synoptic patterns.

Author contributions. C. C. Zhan and M. Xie had the original ideas, designed the research, collected the data, and prepared the original draft. C. C. Zhan carried out the data analysis. M. Xie acquired financial support for the project leading to this publication. C. W. Huang taught and helped C. C. Zhan to do the numerical simulation. T. J. Wang and J. Liu revised the manuscript and helped to collect the data. C. Q. Ma helped to deal with the emission inventory. M. Xu and J. W. Yu helped to collect the data. M. M. Li, S. Li, B. L. Zhuang, and M. Zhao reviewed the initial draft and checked the English of the original manuscript. Y. M. Jiao and D. Y. Nie reviewed the initial draft and helped to improve the work of health impact.

Acknowledgements.

This work was supported by the National Key Research and Development Program of China (2018YFC0213502, 2018YFC1506404), the open research fund of Chongqing Meteorological Bureau (KFJJ-201607) and the Fundamental Research Funds for the Central Universities (020714380047).

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