

1 A Comparative Study to Reveal the Influence of Typhoons on the 2 Transport, Production and Accumulation of O₃ in the Pearl River 3 Delta, China

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14 **Abstract.** The Pearl River Delta (PRD) region in South China is faced with severe ambient O₃ pollution in autumn and summer,
15 which mostly coincides with the occurrence of typhoons above the Northwest Pacific. With increasingly severe O₃ pollution
16 in the PRD under the influence of typhoons, it is necessary to gain a comprehensive understanding of the impact of typhoons
17 on O₃ transport, production and accumulation for efficient O₃ reduction. In this study, we analysed the general influence of
18 typhoons on O₃ pollution in the PRD via systematic comparisons of meteorological conditions, O₃ processes and sources on
19 O₃ pollution days with and without typhoon occurrence (denoted as the typhoon-induced and no-typhoon scenarios,
20 respectively), and also examined the differences in these influences in autumn and summer. The results show that the approach
21 of typhoons was accompanied by higher wind speeds and strengthened downdrafts in autumn as well as the inflows of more
22 polluted air masses in summer, suggesting favourable O₃ transport conditions in the typhoon-induced scenario in both seasons.
23 However, the effect of typhoons on the production and accumulation of O₃ were distinct. Typhoons led to reduced cloud cover,
24 and thus stronger solar radiation in autumn, which accelerated O₃ production, but the shorter residence time of local air masses
25 was unfavourable for the accumulation of O₃ within the PRD. In contrast, in summer, typhoons increased cloud cover, and
26 weakened solar radiation, thus restraining O₃ formation, but the growing residence time of local air masses favoured O₃
27 accumulation. The modelling results using the Community Multiscale Air Quality (CMAQ) model for the typical O₃ pollution
28 days suggest increasing contributions from the transport processes as well as sources outside the PRD for O₃ pollution,
29 confirming enhanced O₃ transport under typhoon influence in both seasons. The results of the process analysis in CMAQ
30 suggest that the chemical process contributed more in autumn but less in summer in the PRD. Since O₃ production and
31 accumulation cannot be enhanced at the same time, the proportion of O₃ contributed by emissions within the PRD was likely
32 to decrease in both seasons. The difference in the typhoon influence on O₃ processes in autumn and summer can be attributed
33 to the seasonal variation of the East Asian monsoon. From the “meteorology-process-source” perspective, this study revealed

34 the complex influence of typhoons on O₃ pollution in the PRD and their seasonal differences. To alleviate O₃ pollution under
35 typhoon influence, emission control is needed on a larger scale, rather than only within the PRD.

36 **1 Introduction**

37 Tropospheric ozone (O₃) serves as a secondary pollutant in ambient air and is detrimental for human health and crop
38 production (Wang et al., 2017; Liu et al., 2018; Mills et al., 2018). Ambient O₃ is produced from its precursors, i.e., nitrogen
39 oxides (NO_x = NO + NO₂) and volatile organic compounds (VOCs), through chemical reactions in the presence of sunlight.
40 Due to the relatively long lifetime of O₃ (~22 days; Stevenson et al., 2006), it can accumulate locally, or be transported to
41 downwind regions. Under unfavourable meteorological conditions, enhanced transport, production and/or accumulation of
42 O₃ can all contribute to the O₃ pollution within a region (National Research Council, 1991).

43

44 As the largest city cluster in South China, the Pearl River Delta (PRD) region is faced with frequent ambient O₃ pollution,
45 especially in autumn and summer (Li et al., 2014; Wang et al., 2017; Lu et al., 2018). Along with the continuous increasing of
46 O₃ levels in recent years (Li et al., 2019), O₃ has become the primary contributor to the deterioration of air quality in this
47 region (Feng et al., 2019). The occurrence of O₃ pollution in the PRD is predominantly related to the influence of typhoons
48 (or tropical cyclones) above the Northwest Pacific (Gao et al., 2018; Deng et al., 2019; Lin et al., 2019). According to Gao et
49 al. (2018), seven out of the nine most severe O₃ episodes (regional-mean maximum 8-h average O₃ concentrations > 240
50 µg/m³) during 2014–2016 coincided with the approach of typhoons. The changes in the track and intensity of typhoons may
51 contribute to the growing trend of O₃ levels recently and in future (Lam, 2018; Lam et al., 2018). Therefore, a
52 comprehensive understanding of the influence of typhoons on the transport, production and accumulation of O₃ has
53 important implications for efficient and strategic O₃ reduction in the PRD.

54

55 Analyses of typhoon-related O₃ episodes in the PRD have been extensively reported in previous publications. The effect of
56 typhoons on O₃ pollution is closely linked to meteorological conditions that are conducive to the transport, production and/or
57 accumulation of O₃. Stagnation caused by typhoons, characterised by low wind speeds, has been reported during many
58 episodes, and it promotes the accumulation of locally formed O₃ within the PRD (Wang et al., 1998; So and Wang, 2003;
59 Wang and Kwok, 2003; Huang et al., 2005; Lam et al., 2005; Jiang et al., 2008; Zhang et al., 2014; Chow et al., 2019).
60 Strong north or west winds were observed or simulated during several episodes, suggesting the potentially strengthened
61 transport of pollutants under typhoon influence (Wang et al., 2001; Yang et al., 2012; Wang et al., 2015; Wei et al., 2016).
62 Downdrafts on the outskirts of typhoons may promote downward O₃ transport and contribute to near-ground O₃ pollution as
63 well (Lam, 2018), but its appearance in the PRD has only been examined in a few studies. Cloudless conditions and strong
64 solar radiation enhance O₃ production, which is another important cause of O₃ pollution (Wang et al., 1998; Wang and
65 Kwok, 2003; Li et al., 2018; Yue et al., 2018; Chow et al., 2019). In a more direct way, several studies have utilised

66 chemical transport models, along with the Process Analysis (PA) tool and source apportionment (SA) methods, to quantify
67 and compare the contributions of various O₃ processes (e.g., transport and the chemical process) and sources (e.g., local
68 emissions, outside emissions and background) during these episodes. Based on reports by Huang et al. (2005), Lam et al.
69 (2005), Jiang et al. (2008), Wang et al. (2010), Li (2013), Wang et al. (2015), Wei et al. (2016) and Chen et al. (2018),
70 horizontal/vertical transport and chemical production may both be the main contributing process for typhoon-induced O₃
71 pollution in different parts of the PRD. The SA results revealed that emissions within the PRD contributed 40–80% of O₃
72 during typhoon-related O₃ episodes (Li et al., 2012; Li, 2013; Chen et al., 2015), suggesting the potentially important role of
73 O₃ accumulation for O₃ pollution here. However, despite massive episode-based studies, several important questions still
74 remain: Are O₃ transport, production and accumulation within the PRD all enhanced at the same time by typhoons? Do both
75 O₃ pollution seasons (autumn and summer) experience similar impact of typhoons on O₃ pollution? More thorough
76 investigations are needed to answer these questions.

77

78 In this study, we present systematic comparisons between O₃ pollution in the typhoon-induced and no-typhoon scenarios
79 (definitions given in Sect. 2.2) to elucidate the influence of typhoons on O₃ transport, production and accumulation in the PRD
80 and to reveal their seasonal differences. October and July in 2014–2018 were selected as the representative months for autumn
81 and summer, respectively. Multiple datasets, including the ERA-Interim re-analysis, the routine monitoring datasets,
82 trajectories calculated by the Hysplit model and the modelling results of typical O₃ pollution days using the Community
83 Multiscale Air Quality (CMAQ) model, were used in the comparisons. A detailed introduction of these datasets is presented
84 in Sect. 2. The comparisons were conducted from the perspectives of meteorological conditions (Sect. 3), O₃ processes and
85 sources (Sect. 4), and the conclusions about the influence of typhoons on the causes of ambient O₃ pollution in the PRD in the
86 two seasons are illustrated in Sect. 5.

87 **2 Methods**

88 **2.1 Datasets**

89 The detailed information for the datasets utilised in the comparison of meteorological conditions is presented below:

- 90 • **Re-analysis datasets:** We mainly used the ERA-Interim re-analysis product in the ~~analyses-comparisons~~ due to its
91 more available parameters ~~than other re-analysis datasets~~ and high spatial coverage (available at
92 <https://www.ecmwf.int/en/forecasts/datasets/reanalysis-datasets/era-interim>, last accessed: March 2020; Dee et al.,
93 2011; Berrisford et al., 2011). Specifically, meteorological parameters used in the comparisons include the following
94 three categories: (1) near-surface parameters from the ~~analysis-fields~~ **analyses data**, including air temperature (at a
95 height of 2 m), relative humidity (RH, at 1000 hPa), horizontal wind speeds (at a height of 10 m; zonal and meridional
96 wind speeds were also involved in the comparisons), and low (for the height at which pressure/surface pressure > 0.8),
97 medium (for the height at which 0.45 < pressure/surface pressure < 0.8), high (for the height at which pressure/surface

98 pressure < 0.45) and total cloud covers; (2) near-surface parameters from the forecast ~~fields~~data, including plenary
99 boundary layer (PBL) height and net surface solar radiation; and (3) upper air parameters at multiple heights, including
100 horizontal and vertical wind speeds, cloud water content and O₃ mixing ratio. The focus of this study is O₃ pollution
101 during the daytime, and therefore, only the parameters at 14:00 local time (LT) were selected for the analyses (except
102 for net surface solar radiation, which was averaged within 8:00–17:00 LT).

- 103 • **Surface meteorological routine monitoring datasets:** The routine monitoring meteorological data collected at 29
104 national meteorological sites within the PRD (locations shown in Fig. S1a) were also used to explore the
105 meteorological features under the impact of typhoons. The parameters include air temperature, RH, and wind speed and
106 direction (also transformed to zonal and meridional wind speeds in the comparisons) at 14:00 LT.
- 107 • **Typhoon information:** The times, locations and intensities of typhoons were provided by the Chinese Meteorological
108 Administration Best Track Dataset of tropical cyclones (Ying et al., 2014). The tracks of all typhoons that potentially
109 contributed to O₃ pollution in the PRD during the study period (October and July in 2014–2018) are shown in Fig. S2
110 and S3.
- 111 • **O₃ concentrations:** Hourly O₃ concentration data, which were originally released by the China National Environmental
112 Monitoring Centre, were downloaded from <http://beijingair.sinaapp.com> (last accessed: Dec. 2018). Based on the
113 hourly data, we calculated the maximum 1-hr concentrations (MDA1) and maximum 8-hr average concentrations
114 (MDA8) of O₃ in nine municipalities in the PRD (including Guangzhou, Shenzhen, Zhuhai, Foshan, Jiangmen,
115 Zhaoqing, Huizhou, Dongguan and Zhongshan) to identify O₃ pollution days that served as samples in the comparisons.

116 2.2 Definition and classification of O₃ pollution days

117 In this study, O₃ pollution days were defined as the days when the MDA1 exceeds 200 µg/m³ or the MDA8 exceeds 160
118 µg/m³ for O₃ (both are the Grade-II thresholds of the Chinese National Ambient Air Quality Standard (NAAQS), GB 3095-
119 2012) in any of the nine municipalities in the PRD. According to these criteria, there were 78 and 55 O₃ pollution days
120 (given in Table S1 and S2) during October and July in 2014–2018, respectively. The information about these O₃ pollution
121 days in the two representative months is listed in Table 1 (overall) and S3 (monthly), including the numbers of days, their
122 proportions in the month, and the corresponding mean O₃ concentrations (MDA8 and MDA1, highest values among nine
123 municipalities in the PRD). Although there were more O₃ pollution days in October than in July, O₃ pollution under typhoon
124 influence occurred on ~30% days of both months. ~~Higher O₃-MDA1 and MDA8 values can be found with the appearance of
125 typhoons in comparison with days without typhoons in July, whereas these values are similar in October, indicating the
126 important role of typhoons in O₃ pollution in the PRD. Typhoons tend to result in more severe O₃ pollution in the PRD, as
127 indicated by generally higher O₃ MDA1 and MDA8 values with the influence of typhoons than these without typhoons.~~

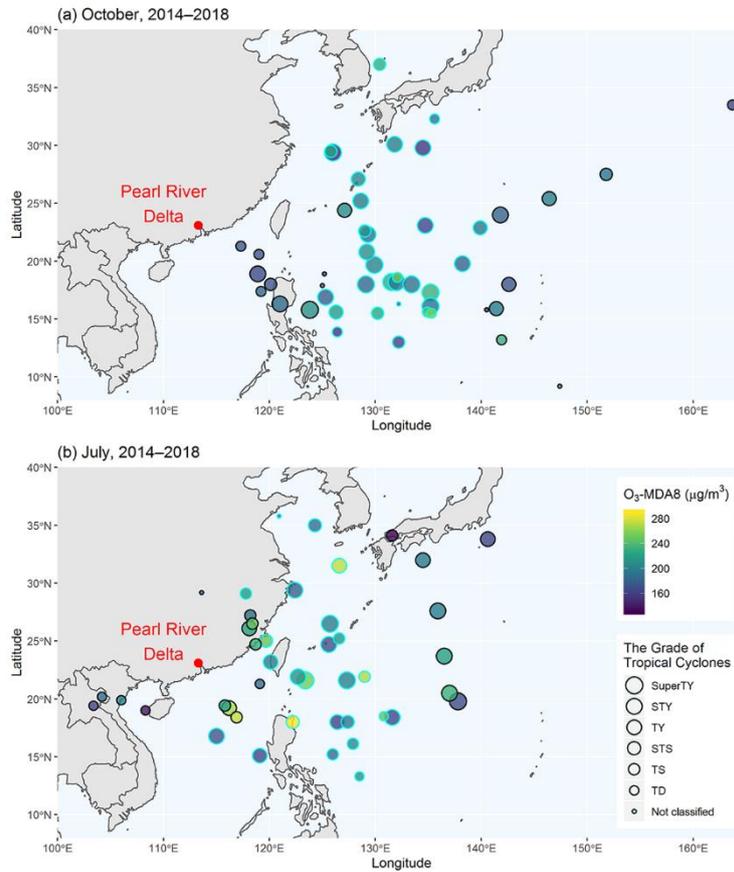
128
129 The differing locations of typhoons can result in the diverse effect of typhoons on O₃ pollution (Chow et al., 2018). To
130 determine the general influence of typhoons on O₃ pollution in the PRD, it was necessary to further select O₃ pollution days

131 coinciding with typhoons with similar directions and distances to the PRD. As is shown in Fig. 1, all O₃ pollution days in
 132 October and most O₃ pollution days in July under typhoon influence were associated with typhoons to the east of the PRD,
 133 which were more likely to cause O₃ pollution (Chow et al., 2018). In order to minimize the disturbance of typhoon directions
 134 in the comparisons, we removed the remaining five O₃ pollution days in July with typhoons located to the due north or
 135 southwest of the PRD from the analyses. After this, based on the distances between typhoon centres and the PRD (at 14:00
 136 LT), we classified the pollution days with typhoons in each season into three categories: close typhoon (lowest 20% of
 137 distances), typhoon (20–80% intervals of distances), and far typhoon (longest 20% of distances)-induced days. The typhoon-
 138 induced days represent O₃ pollution days with general typhoon influence, and they were compared with those without the
 139 appearance of typhoons (hereafter denoted as the no-typhoon days). It should be noted that the distances between typhoon
 140 centres and the PRD on the typhoon-induced days were overall larger in autumn (1400–2800 km, at 14:00 LT) than in
 141 summer (700–2000 km, at 14:00 LT), which may be the consequence of the different characteristics of typhoon paths in the
 142 two seasons: most typhoons in autumn travel northwest initially and then turn northward in the areas east of the Philippines
 143 (Fig. S2), whereas they are more likely to end up landing in Southeast China in summer (Fig. S3). Since the influence of
 144 typhoons on O₃ pollution may be different when typhoons come close enough to the PRD (Lam et al., 2005; Li, 2013), the
 145 close typhoon-induced days were considered to be a special scenario in the comparisons of meteorological conditions (Sect.
 146 3.5). Owing to the less apparent effect of typhoons over the PRD, we did not include the far typhoon-induced days in the
 147 discussions.

148

149 **Table 1.** The numbers and proportions of O₃ pollution days, and O₃ concentrations for various scenarios.

Parameter	October, 2014–2018	July, 2014–2018	150
Number (proportion) of O ₃ pollution days	78 (50.3%)	55 (35.5%)	151
With typhoons	49 (31.6%)	45 (29.0%)	
Typhoon-induced days	30 (19.4%)	24 (15.5%)	
Close typhoon-induced days	10 (6.5%)	8 (5.2%)	
Without typhoons (no-typhoon days)	29 (18.7%)	10 (6.5%)	
Mean PRD-max O ₃ MDA8 (µg/m ³)			
With typhoons	195.0	205.3	
Typhoon-induced days	199.5	205.4	
Close typhoon-induced days	184.6	225.7	
Without typhoons (no-typhoon days)	189.8	187.8	
Mean PRD-max O ₃ MDA1 (µg/m ³)			
With typhoons	230.4	259.8	
Typhoon-induced days	235.2	260.0	
Close typhoon-induced days	219.2	277.1	
Without typhoons (no-typhoon days)	231.5	246.5	



152

153 **Figure 1.** The location and intensity of typhoons at 14:00 LT on all O₃ pollution days with typhoons, and the corresponding O₃ MDA8
 154 concentrations (maximum values in the nine municipalities of the PRD) on the same days during (a) October and (b) July in 2014–2018.
 155 The points with cyan borders indicate the “typhoon-induced” O₃ pollution days used in the comparisons. The grades of tropical cyclones
 156 (Chinese National Standard, GB/T 19201-2006) are as follows: SuperTY - super typhoon; STY - severe typhoon; TY - typhoon; STS -
 157 severe tropical storm; TS - tropical storm; TD - tropical depression; others are grouped as “not classified”.

158 2.3 Calculation of the trajectories and air parcel residence time

159 To explore the potential effect of cross-regional transport on O₃ pollution in the PRD, we applied the Hysplit model (Stein et
 160 al., 2015) with the Global Data Assimilation System (GDAS) datasets as inputs to calculate 72-h backward trajectories reaching
 161 the PRD at 14:00 LT for all O₃ pollution days. The Modiesha site (23.1°N, 113.3°E; Fig. S1b), which is located in the central
 162 part of the PRD, was the endpoint of backward trajectories. Its height was set as 500 m above the ground to better represent
 163 the effect of long-range transport on O₃ pollution, as well as to minimize the disturbance of objects near the surface to the
 164 transport (Park et al., 2007). Its height was set to 500 m above the ground to indicate the effect of long-range transport on O₃
 165 pollution within the PBL (Park et al., 2007).

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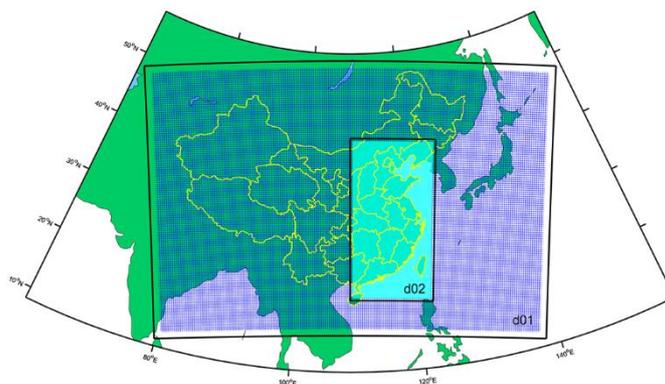
167 Air parcel residence time (APRT), discussed by Huang et al. (2019), is the average number of hours that air parcels originated
168 from one place stay within a pre-defined domain, and long APRTs can be used to indicate good accumulation conditions for
169 locally sourced pollutants. To calculate APRTs in the PRD, we designed a 21×15 point matrix (resolution: 0.2°×0.2°) that
170 embraces the whole PRD (Fig. S4), and forward trajectories starting from these points were also calculated using the Hysplit
171 model. The height of all points was set as 100 m above the ground to represent the height of all local emissions and to reduce
172 the disturbance of the surface, as well. The start times were set as 2:00, 8:00, 14:00 and 20:00 LT for all O₃ pollution days.
173 Afterwards, the length of time each trajectory remained within the administration borders of the PRD, i.e., APRT, was
174 calculated and attributed to its starting point. APRTs in each point were averaged, and these averaged APRT values in all
175 points were interpolated using the Kriging method to obtain field results for the further comparisons.

176 2.4 CMAQ modelling: basic setups and modelling methods

177 We utilised the widely used 3D chemical transport model CMAQ (version 5.0.2) to investigate the effects of typhoons on O₃
178 processes and sources. October 2015 and July 2016 featured the most severe O₃ pollution under typhoon influence among all
179 representative months ~~of the two seasons in autumn and summer, respectively~~ (Table S3), ~~and thus, they were chosen as the~~
180 ~~period in the modelling (because there was no severe O₃ pollution during the first 10 days of October 2015 and 3–5 November~~
181 ~~can be classified as the no-typhoon O₃ pollution days, we adjusted the modelling period in autumn to 11 October–10 November~~
182 ~~2015) and all typhoon-induced and no-typhoon O₃ pollution days in these two months served as representative O₃ pollution~~
183 ~~days in the comparisons. Therefore, O₃ pollution in these two months were modelled and used in further comparisons. Note~~
184 ~~that the modelling period in autumn was adjusted to 11 October–10 November 2015 to involve more O₃ pollution days (3–5~~
185 ~~November 2015).~~ In detail, there were four typhoon-induced O₃ pollution days (14–16 and 21 October 2015) and four no-
186 typhoon O₃ pollution days (28 October and 3–5 November 2015) in October 2015, whereas there were four and six typhoon-
187 induced and no-typhoon days in July 2016, respectively (typhoon-induced: 7–8 and 30–31 July 2016; and no-typhoon: 22–26
188 and 29 July 2016). The results of daytime (9:00–17:00 LT) O₃ PA and SA on the above O₃ pollution days were averaged for
189 the typhoon-induced and no-typhoon scenarios in autumn (October 2015) and summer (July 2016) and were used in the
190 comparisons.

191
192 The main setups of the CMAQ model are presented as follows. Two-nested modelling domains with the resolutions of 36 and
193 12 km (denoted as d01 and d02, respectively) were set in this study (Fig. 2). Specifically, d02 covers the whole East and
194 Central China (EC-China), enabling us to evaluate the contribution of emissions in these areas to O₃ pollution in the PRD.
195 There were 19 vertical layers in the CMAQ modelling, with about 10 layers within the PBL (about 0–1 km in heights; Guo et
196 al., 2016). The Weather Research and Forecasting (WRF) model (version 3.2) provided the meteorological fields used as inputs.
197 SMOKE (version 2.5) and MEGAN (version 2.10) were used to process the anthropogenic and biogenic emissions ~~files,~~
198 respectively. The anthropogenic emission inventory used in this study consisted of the following three parts: (1) emissions in
199 the PRD, which were provided by the Guangdong Environmental Monitoring Centre; (2) emissions in other areas of mainland

200 China, which were extracted from the MEIC inventory (He, 2012); and (3) emissions in other countries and regions in Asia,
201 which were extracted from the MIX inventory (Li et al., 2017). The initial and boundary conditions of the d01 modelling were
202 obtained from the same-period results of the MOZART-4 global model (available at <https://www.acom.ucar.edu/wrf-chem/mozart.shtml>, last accessed: Dec. 2019), and those of the d02 modelling were extracted from the d01 modelling results.
203 The SAPRC07 gas-phase chemistry mechanism (Carter, 2010) and the AERO6 aerosol scheme were set in the modelling. In
204 addition, the simulations of the two months were both started 10 days ahead to minimise the disturbance of the bias of the
205 initial conditions. The modelling performances of CMAQ and WRF were determined to be acceptable based on the
206 comparisons between the observational and modelling series of meteorological parameters, O₃ MDA8, daily NO₂
207 concentrations and the mixing ratios of non-methane hydrocarbons (NMHCs) in the PRD (for details, refer to Sect. 1 in the
208 Supplement Information), which ensures the validity of the further analyses.
209
210



211
212 **Figure 2.** Two-nested modelling domain, noted as d01 and d02. The black boxes indicate the WRF modelling domains, and the nested
213 areas are the CMAQ modelling domains.

214 The PA tool in CMAQ was implemented to quantify the hourly contributions of O₃ processes (or integrated process rate, IPR),
215 which includes vertical/horizontal transport (convection+diffusion), chemical process (net O₃ production through gas-phase
216 reactions), dry deposition and cloud process. To explore the overall effect of typhoons on O₃ transport and production in the
217 region, the mean PA results within the administration boundaries of the PRD were calculated and compared.
218

219 In order to identify the sources of all O₃ in the PRD, we used the classic Brute Force Method (BFM) to identify the contributions
220 of emissions (including anthropogenic and biogenic emissions) in the PRD and other regions in the d02 (mainly EC-China),
221 as well as regions outside the d02 (the boundary conditions of the d02) for O₃ pollution in the PRD (hereafter denoted as the
222 contributions of PRD, EC-China and BCON, or S_{PRD} , $S_{EC-China}$ and S_{BCON} , respectively). For a pollutant, the contribution of a
223 specific emission, E_i , can be calculated in two ways: (1) the difference between the modelled concentrations of the base case
224 (all emissions involved) and the sensitivity case where E_i is zeroed out (i.e., top-down BFM); (2) the difference between two
225 sensitivity cases where emissions ~~except~~except E_i and all of the emissions are zeroed out, respectively (i.e., bottom-up BFM).

226 Owing to the non-linearity between O₃ and its precursors, biases may occur between the results of two types of BFM methods,
227 leading to the non-additivity of the results (Clappier et al., 2017). Therefore, the average of the top-down and bottom-up BFM
228 results was treated as the quantified contributions of the concerned sources. Four simulation cases were run in this study,
229 including (the modelled O₃ concentration in each case was also marked in brackets):

- 230 • the base case (C_{base});
- 231 • the PRD-cut case (C_{PRD_cut}), where emissions within the PRD were zeroed out;
- 232 • the PRD-only case (C_{PRD_only}), where emissions outside the PRD (within d02) were zeroed out; and
- 233 • the zero-emission case (C_0), where all emissions within the d02 were zeroed out.

234 Afterwards, the S_{PRD} , $S_{EC-China}$ and S_{BCON} values (in concentrations) in the polluted areas of the PRD (where modelled daytime
235 O₃ concentrations > 160 µg/m³, the Grade-II O₃ MDA8 thresholds of the Chinese NAAQS) were calculated using the following
236 equations,

$$S_{PRD} = \frac{1}{2} [(C_{base} - C_{PRD_cut}) + (C_{PRD_only} - C_0)], \quad (R1)$$

$$S_{EC-China} = \frac{1}{2} [(C_{base} - C_{PRD_only}) + (C_{PRD_cut} - C_0)], \quad (R2)$$

$$S_{BCON} = C_0. \quad (R3)$$

237 The percentage forms of these values were used in the comparisons.

238 3 Comparison of meteorological conditions

239 3.1 Overview: comparison of meteorological parameters in the PRD

240 First, we compared near-ground meteorological parameters in the PRD on the typhoon-induced and no-typhoon O₃ pollution
241 days. ~~The parameters from routine monitoring datasets (including air temperature, RH, wind speed, zonal and meridional~~
242 ~~wind speeds measured at 14:00 LT of all O₃ pollution days at 29 national meteorological sites within the PRD (Fig. S1a))~~
243 ~~and the ERA-Interim re-analysis (including all near-surface parameters from the analysis and forecast fields introduced in~~
244 ~~Sect. 2.1, extracted at the same time and the locations of sites as these in routine monitoring datasets) were used in the~~
245 ~~comparison (since all O₃ pollution days in October and over 60% of O₃ pollution days in July were characterized with sunny,~~
246 ~~cloudy, or overcast weathers with no rainfall in the PRD (Table S4, represented by the weather in Guangzhou), precipitation~~
247 ~~was not considered in the comparisons). Near-surface parameters from routine monitoring datasets and the ERA-Interim re-~~
248 ~~analysis were used in the comparison. The parameters include air temperature, RH, wind speeds, cloud covers, PBL height~~
249 ~~and net surface solar radiation. Since there was no rainfall on most O₃ pollution days (indicated by the weather in Guangzhou~~
250 ~~(Table S4)), precipitation was not considered in the comparisons. For consistency, the parameters of ERA-Interim were~~
251 ~~extracted at the same time (14:00 LT) and the locations of sites (Fig. S1a) as those in routine monitoring.~~ The Mann-Whitney
252 U test was applied to determine whether the above parameters were significantly different ($p < 0.05$) between typhoon-
253 induced and no-typhoon O₃ pollution days.

255 As is listed in Table 2, statistically significant differences between the typhoon-induced and no-typhoon scenarios existed for
 256 most of the parameters, such as meridional (south-north) wind speed, cloud covers within various height ranges and net
 257 surface solar radiation — in both seasons, these parameters were significantly different for the two scenarios. It indicates that
 258 the causes of O₃ pollution may vary on typhoon-induced and no-typhoon O₃ pollution days. Note that air temperature, one of
 259 the parameters most closely related to O₃ pollution in the PRD (Zhao et al., 2019), was not significantly different in the two
 260 scenarios. We also found that the comparison in autumn and summer did not produce the same results: the typhoon-induced
 261 days in autumn featured lower RH, stronger winds (especially north wind), reduced cloud cover (low, medium, high and
 262 total) and stronger surface solar radiation, whereas in summer, these days had higher RH, weaker south winds, more cloud
 263 cover (medium, high and total), weaker surface solar radiation and lower PBL heights. Therefore, the impact of typhoons on
 264 O₃ pollution differs in the two seasons, as well. In order to reveal the impact of typhoons on O₃ transport, production, and
 265 accumulation in the PRD, more detailed comparisons of the corresponding meteorological indicators are presented in the
 266 following sections.

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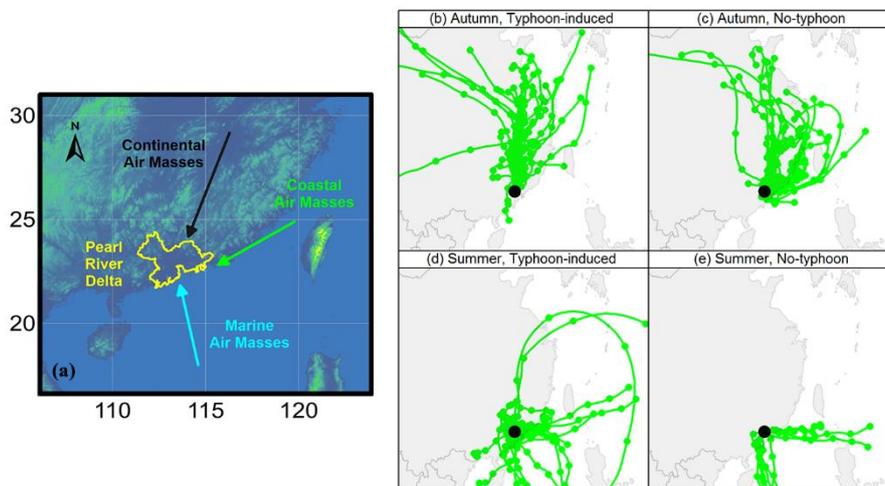
268 **Table 2.** The comparisons of meteorological parameters (all at 14:00 LT except for net surface solar radiation, which is the average value
 269 for 9:00–17:00 LT) in the PRD for the three scenarios (no-typhoon, typhoon-induced, close typhoon-induced) in two seasons (autumn,
 270 summer). RM, routine measurement; ERA, ERA-Interim re-analysis. All of the parameters are presented as “the mean value ± standard
 271 deviation”. The differences between parameters in the typhoon-induced or close typhoon-induced scenarios and the corresponding
 272 typhoon-induced scenarios for the same season are given in parentheses, and “*” indicates $p < 0.05$, or statistically significant differences
 273 between these parameters when the Mann-Whitney U test is used.

Parameters	Data Source	Autumn (October, 2014–2018)			Summer (July, 2014–2018)		
		No-typhoon	Typhoon-induced	Close Typhoon-induced	No-typhoon	Typhoon-induced	Close Typhoon-induced
Air Temperature (°C)	RM	29.1 ± 2.2	29.3 ± 1.8 (+0.2)	29.6 ± 1.5 (+0.5, *)	33.7 ± 2.0	33.9 ± 2.0 (+0.2)	35.0 ± 1.5 (+1.3, *)
	ERA	29.2 ± 2.1	29.3 ± 1.6 (+0.1)	29.6 ± 1.5 (+0.4, *)	33.4 ± 1.8	33.5 ± 1.4 (+0.1)	34.6 ± 1.4 (+1.2, *)
RH (%)	RM	52.4 ± 10.2	44.8 ± 10.4 (-7.6, *)	51.4 ± 12.4 (-1.0)	57.0 ± 9.3	58.3 ± 9.7 (+1.3)	56.9 ± 6.4 (-0.1)
	ERA	54.0 ± 9.8	48.3 ± 11.2 (-5.7, *)	52.2 ± 12.4 (-1.8, *)	62.6 ± 10.8	66.4 ± 9.4 (+3.8, *)	62.5 ± 9.4 (+0.1)
Wind Speed (m/s)	RM	2.33 ± 1.18	2.58 ± 1.23 (+0.25, *)	2.96 ± 1.40 (+0.63, *)	2.46 ± 1.33	2.30 ± 1.20 (-0.16)	2.53 ± 1.16 (+0.07)
	ERA	2.39 ± 1.30	2.54 ± 0.99 (+0.15, *)	3.53 ± 1.11 (+1.14, *)	2.41 ± 0.99	2.18 ± 1.18 (-0.23, *)	2.61 ± 1.05 (+0.20)
Zonal (East-West) Wind Speed (m/s)	RM	-0.83 ± 1.72	-0.59 ± 1.70 (+0.24, *)	-0.13 ± 1.74 (+0.70, *)	-0.41 ± 2.05	-0.03 ± 1.94 (+0.38)	0.73 ± 1.98 (+1.14, *)
	ERA	-1.41 ± 1.43	-1.07 ± 1.04 (+0.34, *)	-0.87 ± 0.79 (+0.54, *)	0.22 ± 1.73	-0.02 ± 1.81 (-0.24)	0.29 ± 2.45 (+0.07)
Meridional (South-North) Wind Speed (m/s)	RM	-0.36 ± 1.74	-1.49 ± 1.66 (-1.13, *)	-2.21 ± 1.66 (-1.85, *)	0.79 ± 1.69	0.01 ± 1.72 (-0.78, *)	-0.69 ± 1.68 (-1.48, *)
	ERA	-0.27 ± 1.82	-1.97 ± 1.16	-3.27 ± 1.29	1.61 ± 1.09	0.64 ± 1.58	-0.68 ± 1.19

			(-1.70, *)	(-3.00, *)		(-0.97, *)	(-2.29, *)
Low Cloud	ERA	17.2 ± 22.7	4.2 ± 11.9	15.5 ± 23.9	8.7 ± 9.4	7.1 ± 9.5	5.2 ± 5.0
Cover (%)			(-13.0, *)	(-1.7, *)		(-1.6, *)	(-3.5, *)
Medium Cloud	ERA	22.2 ± 26.5	10.4 ± 19.7	9.5 ± 14.5	8.7 ± 11.1	15.4 ± 15.1	21.5 ± 15.5
Cover (%)			(-11.8, *)	(-12.7, *)		(+6.7, *)	(+12.8, *)
High Cloud	ERA	12.1 ± 23.1	7.2 ± 16.3	34.6 ± 35.6	32.2 ± 30.0	44.9 ± 29.3	51.0 ± 34.2
Cover (%)			(-4.9, *)	(+22.5, *)		(+12.7, *)	(+18.8, *)
Total Cloud	ERA	43.5 ± 32.3	20.5 ± 25.7	51.9 ± 33.1	43.7 ± 26.7	58.3 ± 22.7	67.5 ± 21.0
Cover (%)			(-23.0, *)	(+8.4, *)		(+14.6, *)	(+23.7, *)
Net Surface Solar	ERA	456.9 ± 78.4	516.6 ± 66.7	516.5 ± 62.8	560.3 ± 93.1	523.2 ± 74.4	541.9 ± 54.0
Radiation (W/m ²)			(+59.7, *)	(+59.6, *)		(-37.1, *)	(-18.4, *)
PBL Height (m)	ERA	1471 ± 315	1473 ± 348	1349 ± 227	1268 ± 383	1037 ± 289	1196 ± 300
			(+2)	(-122, *)		(-231, *)	(-72, *)

274 3.2 O₃ transport conditions: comparison of wind speeds, backward trajectories and vertical air motions

275 The higher wind speeds and/or O₃ levels in the transported air masses are, the more likely O₃ transport plays an increasingly
 276 important role in O₃ pollution. In the PRD, O₃ levels are closely linked to the type of air masses influencing the region, which
 277 can be identified based on backward trajectories. According to Zheng et al. (2010), there are generally three types of air masses
 278 that are transported into the PRD along different paths and contribute to O₃ pollution here, namely, the continental, coastal and
 279 marine air masses (Fig. 3a). The continental and coastal air masses can bring O₃ from EC-China to the PRD, and thus, they
 280 are typically recognised as being polluted and contributing to relatively high O₃ levels in the PRD. In contrast, the marine air
 281 masses, originated from the South China Sea, are much cleaner. In this section, we studied the influence of typhoons on O₃
 282 transport by comparing wind speeds and 72-h backward trajectories in the typhoon-induced and no-typhoon scenarios.
 283



284
 285 **Figure 3.** (a) Three O₃ transport paths towards the PRD. (b–e) Backward trajectories at 14:00 LT for each scenario: (b) autumn, typhoon-
 286 induced; (c) autumn, no-typhoon; (d) summer, typhoon-induced; and (e) summer, no-typhoon. The black dots indicate the end point of all
 287 trajectories, i.e., where the Modiesha site in the central PRD is located.

288 As is displayed in Fig. 3b–c, we identified the influence of continental air masses on the typhoon-induced O₃ pollution days
289 in autumn, as well as mixed contributions from the continental and coastal air masses on the corresponding no-typhoon days.
290 However, for the former scenario, significantly increased wind speeds (Table 2) ensure more favourable conditions for the
291 transport of O₃. In summer, the three types of air masses may all have affected O₃ pollution in the typhoon-induced scenario,
292 while only the marine air masses influenced the PRD in the no-typhoon scenario (Fig. 3d–e). The increasing influence of much
293 more polluted air masses (continental and coastal air masses) led by typhoon ensured that more O₃ was transported to the PRD,
294 thus typhoons also tended to increase the contribution of transport to O₃ pollution in the PRD in summer. In addition, the
295 influence of different air masses was also accompanied with variations in the prevailing winds in the PRD, that is, north winds
296 and easterlies in the typhoon-induced and no-typhoon scenarios in autumn, respectively, and ~~southwest~~southerly-winds in the
297 no-typhoon scenario in summer (indicated by wind roses in Fig. S5). For the typhoon-induced scenario in summer, the
298 dominate wind direction is hard to determine. These variations in the local wind fields potentially result in the different spatial
299 distribution of O₃ concentrations in various scenarios.

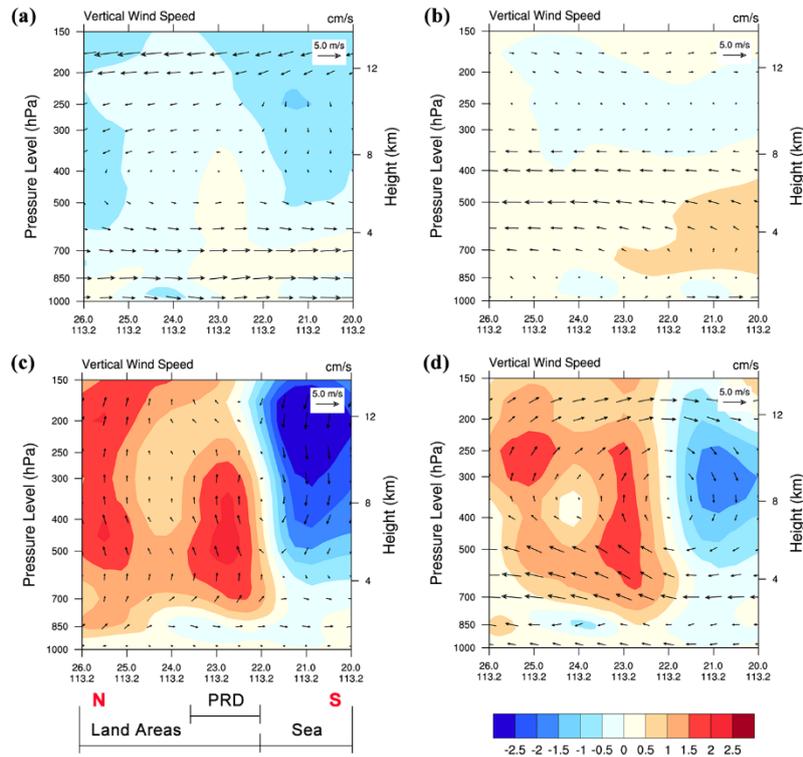
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301 Downdrafts are typically considered to be an important cause of typhoon-induced O₃ pollution (Lam, 2018), but in which
302 scenarios downdrafts influence the PRD remains unclear. Thus, we explored the overall features of vertical air motions from
303 the surface layer to the tropopause in the typhoon-induced and no-typhoon scenarios, and the ERA-Interim reanalysis dataset
304 (including all upper air parameters at multiple heights introduced in Sect. 2.1) was utilised in the comparisons. The contours
305 in Fig. 4 show the cross sections of mean vertical wind speed at 14:00 LT of all O₃ pollution days corresponding to the typhoon-
306 induced and no-typhoon scenarios of two seasons, which were made along the 113.2°E longitude line, from 26.0°N to 20.0°N
307 (Fig. S4). On the typhoon-induced days in autumn, downdrafts occurred over large areas above the PRD, especially above a
308 height of ~700 hPa. Although updrafts can still be found near the sea surface in this scenario, vertical wind speeds tended to
309 be lower compared with those on the no-typhoon days in autumn, which also suggests the enhancement of downdrafts caused
310 by typhoons. In summer, the influence of downdrafts was found over the PRD under 850 hPa on the typhoon-induced O₃
311 pollution days. However, overall, updrafts prevailed above the land areas and downdrafts prevailed above the sea in both the
312 typhoon-induced and no-typhoon scenarios in summer, which is recognised as the structure of the East Asian summer monsoon
313 cell (Chen et al., 1964; Jin et al., 2013; Ding et al., 2018). For both updrafts and downdrafts, the absolute values of vertical
314 wind speeds in the typhoon-induced scenario in summer were overall higher than ~~these~~those in the corresponding no-typhoon
315 scenario. Therefore, the approach of typhoons did not break the structure of the summer monsoon cell, but rather they further
316 strengthened the vertical motions above both land areas and sea. These analyses suggest that typhoons do not necessarily lead
317 to downdrafts during O₃ pollution periods in the PRD and its adjacent areas; and in summer, vertical air motions affected by
318 typhoons are more complicated than expected owing to the existence of the East Asian summer monsoon.

319

320 We also explored the regions where downdrafts and updrafts occurred on a larger scale and their potential connections with
321 O₃ levels. As is shown in Fig. 5, though updrafts appeared in the PRD at 850 hPa on the typhoon-induced days in autumn,

322 downdrafts dominated in the region at 700 and 500 hPa. For the areas to the north of the PRD, the important role of downdrafts
 323 was found at all three heights. In contrast to the no-typhoon days in autumn, downdrafts tended to cover much larger areas in
 324 this scenario. Moreover, these areas at 850 and 700 hPa generally featured higher O₃ mixing ratios as well as lower RH (Fig.
 325 S6) than others, which is a sign of possible direct downward O₃ transport (Roux et al., 2020; Wang et al., 2020). This part of
 326 O₃ can notably aggravate near-ground O₃ pollution in the PRD. In contrast, in summer, updrafts dominated the PRD at various
 327 heights in both scenarios. Besides the PRD, most of the regions near the coast were characterised by updrafts above the land
 328 as well as downdrafts offshore, further indicating the ubiquity of the summer monsoon cell. By comparing the two scenarios
 329 in summer, we found that typhoons resulted in more areas being influenced by updrafts. The areas with high O₃ levels did not
 330 coincide with the downdraft-affected areas, and therefore, O₃ transported from the upper air may play a less significant role in
 331 the typhoon-induced O₃ pollution in summer.



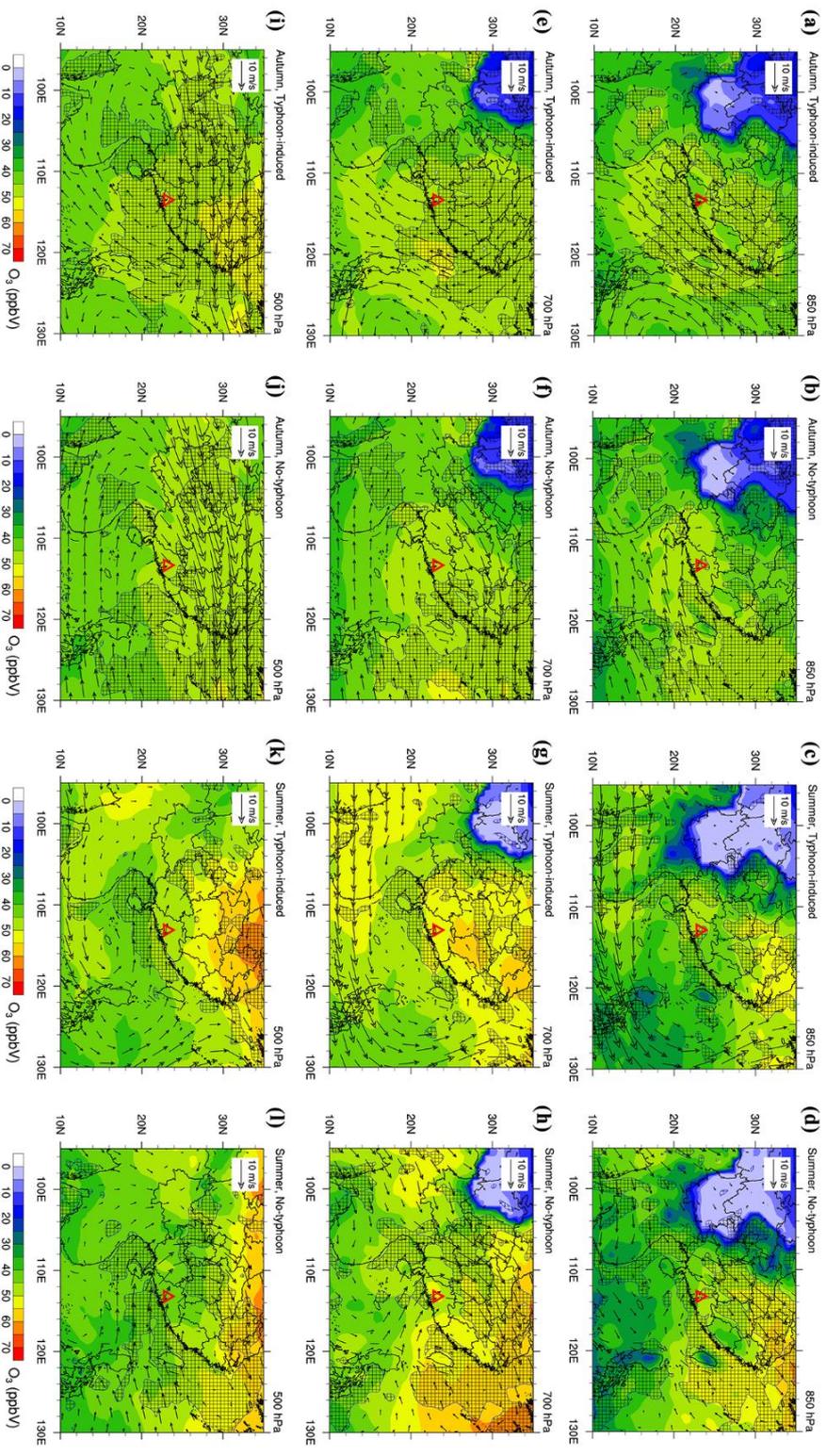
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333 **Figure 4.** The cross sections of mean vertical wind field at 14:00 LT for each scenario: (a) autumn, typhoon-induced; (b) autumn, no-
 334 typhoon; (c) summer, typhoon-induced; and (d) summer, no-typhoon. Cross sections were made along the 113.2°E longitude line, from
 335 26.0°N to 20.0°N (Fig. S4). The vectors indicate meridional wind speed (m/s) and vertical wind speed (cm/s), and the contours indicate
 336 vertical wind speed (cm/s). PRD, the Pearl River Delta.

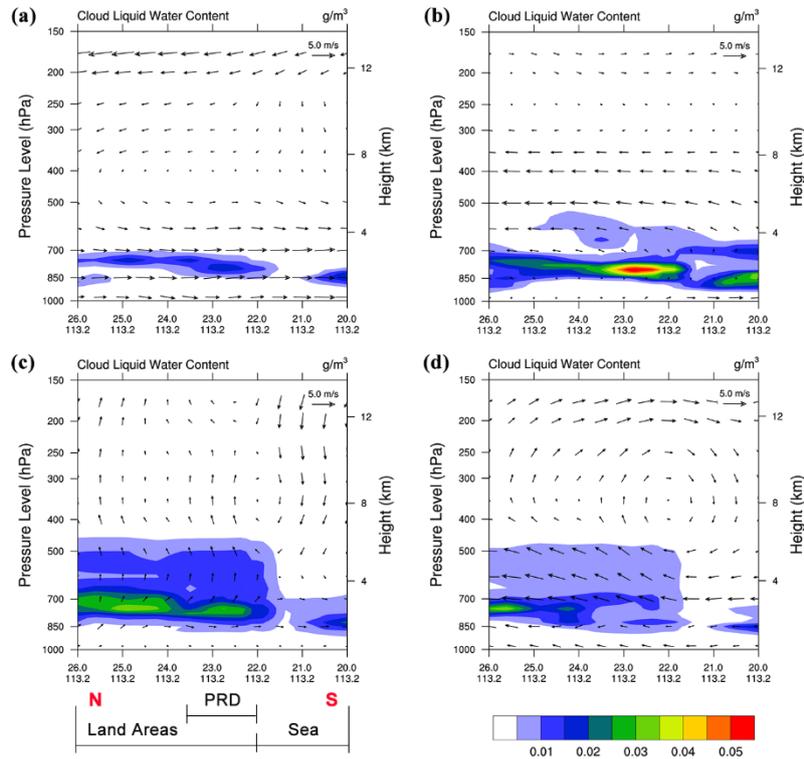
337 3.3 O₃ production conditions: comparison of clouds

338 Clouds efficiently reflect solar radiation (Liou, 1976), and therefore, they have a notable impact on the local formation of O₃.
 339 Figure 6 displays the cross sections of mean ERA-Interim cloud liquid water contents (CLWC) at 14:00 LT of all O₃ pollution

340 days corresponding to the typhoon-induced and no-typhoon scenarios of two seasons, which were also made along the 113.2 E
341 longitude line, from 26.0 N to 20.0 N (Fig. S4). The comparison of CLWC in the cross sections suggests that typhoons
342 generally resulted in fewer clouds in autumn but more clouds in summer, which agrees well with the comparison of cloud
343 covers in Table 2. The presence of fewer clouds on the typhoon-induced days in autumn can be attributed to two reasons: the
344 influence of dry air masses (indicated by lower RH in Table 2 and Fig. S6) and/or the hindrance of cloud formation by
345 downdrafts. In summer, the strengthened updrafts above the land caused by typhoons favoured cloud formation, which is
346 demonstrated by higher CLWC at the heights of 500–850 hPa and increases in medium and high cloud covers. In areas above
347 the PRD below 850 hPa, downdrafts led to slight decrease of clouds in the typhoon-induced scenario in summer, which is also
348 indicated by reduced low cloud cover. As a consequence of varied cloud covers in each scenario, on average, net surface solar



350 **Figure 5.** O₃ mixing ratio (ppbv) and wind fields at the height of (a–d) 850 hPa, (e–h) 700 hPa, and (i–l) 500 hPa at 14:00 LT for each scenario: (a, e, i) autumn,
 351 typhoon-induced; (b, f, j) autumn, no-typhoon; (c, g, k) summer, typhoon-induced; and (d, h, l) summer, no-typhoon. The red triangle in each plot indicates the
 352 PRD. The gridded areas indicate that vertical wind speed is less than 0, or downdrafts occur.



353

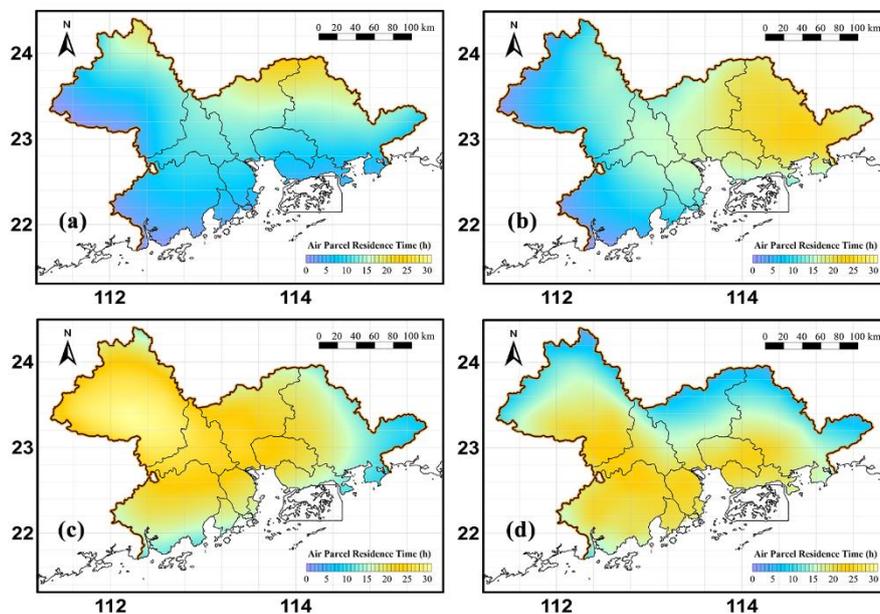
354 **Figure 6.** The cross sections of mean cloud liquid water content (g/m^3) and wind vectors at 14:00 LT for each scenario: (a) autumn, typhoon-
 355 induced; (b) autumn, no-typhoon; (c) summer, typhoon-induced; and (d) summer, no-typhoon. Cross sections were made along the 113.2°E
 356 longitude line, from 26.0°N to 20.0°N (Fig. S4). The vectors indicate meridional wind speed (m/s) and vertical wind speed (cm/s). PRD, the
 357 Pearl River Delta.

358 radiation increased by 13% and decreased by 7% on the typhoon-induced days in autumn and summer, respectively (Table 2),
 359 which promoted and hindered O_3 production in the PRD during these two seasons, respectively.

360 3.4 O_3 accumulation conditions: comparison of APRTs

361 The longer APRTs are, the more likely that O_3 produced by local emissions accumulates within the targeted region and notably
 362 contributes to near-ground O_3 pollution. In order to study the effect of typhoons on O_3 accumulation, we calculated APRTs in
 363 the PRD in the typhoon-induced and no-typhoon scenarios (Fig. 7) for the further comparisons. On the typhoon-induced days
 364 in autumn, APRTs were typically 5–10 hours (mean = 9.5 hours) — shorter than those on the no-typhoon days in autumn
 365 (mean = 13.1 hours). In addition, lower APRT values occurred in the central part of the PRD, where high anthropogenic
 366 emissions of pollutants are distributed (Zheng et al., 2009). Despite more active O_3 chemistry discussed in the last section,
 367 locally sourced O_3 was less likely to accumulate within the PRD in this scenario, potentially limiting the contribution of local
 368 emissions for O_3 . The comparison suggests opposite results in the summer scenarios, that is, APRTs on the typhoon-induced
 369 days (20–30 hours, mean = 21.0 hours) were overall higher than those on the no-typhoon days (15–25 hours, mean = 16.5
 370 hours). This favoured the accumulation of locally sourced O_3 , and, to some extent, offset the influence of weakened O_3

371 ~~formation to maintain high contributions of local emissions to O₃ pollution.~~ Based on the comparisons ~~of O₃ production~~
372 ~~conditions in the previous section and the comparison of O₃ accumulation conditions~~ in this ~~and previous~~ section, typhoons
373 did not provide more favourable conditions for O₃ production and accumulation simultaneously in the PRD in both autumn
374 and summer, thus potentially resulting in ~~a less important role of local reduced~~ contributions ~~in percentage of local emissions~~
375 ~~in to~~ O₃ pollution here. More quantitative evaluations of the contributions from multiple O₃ sources are discussed in Sect. 4.
376



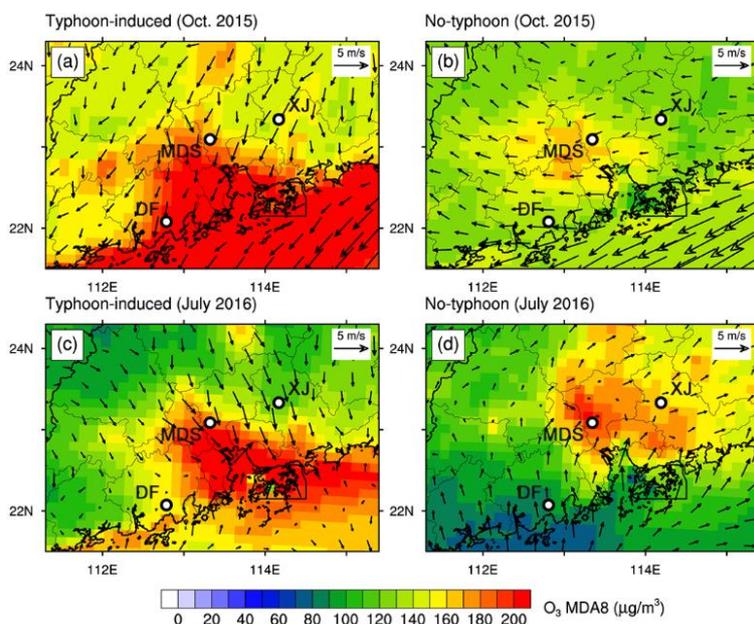
377
378 **Figure 7.** The spatial distributions of APRTs in the PRD for each scenario: (a) autumn, typhoon-induced; (b) autumn, no-typhoon; (c)
379 summer, typhoon-induced; and (d) summer, no-typhoon.

380 3.5 Meteorological conditions on the close typhoon-induced days

381 On the close typhoon-induced days in the two seasons, stronger north winds prevailed and total cloud cover was higher than
382 that on the no-typhoon days (Table 2), suggesting better conditions for the transport of O₃ but less favourable conditions for
383 O₃ production. As displayed in Fig. S7, the APRT values were significantly lower on the close typhoon-induced days (mean
384 = 6.6 hours, 12.9 hours in autumn and summer, respectively) than on the no-typhoon days, making it even harder for locally
385 sourced O₃ to accumulate within the PRD. Therefore, close typhoons are concluded to promote the transport of O₃ from the
386 outside and to reduce the contributions of O₃ produced from local emissions in a more notable way. In addition, close typhoons
387 led to stronger downdrafts in autumn and updrafts in summer than other scenarios in the same season (Fig. S8). It should be
388 noted that the structure of the summer monsoon cell near the PRD was destroyed in the close typhoon-induced scenario in
389 summer, indicating the stronger influence of typhoons on regional wind fields. The dominant role of O₃ transport during O₃
390 pollution days in this special scenario agrees well with the reported episode-based analyses (Lam et al., 2005; Li, 2013).

391 4 Comparisons of O₃ processes and sources

392 The comparisons of meteorological conditions served as qualitative evidence to determine the general influence of typhoons
393 on O₃ transport, production and accumulation in autumn and summer. Based on the comparison between the CMAQ modelling
394 results on typical O₃ pollution days in October 2015 and July 2016, more quantitative evidence can be presented. Figure 8
395 displays modelled mean O₃ MDA8 concentrations and wind fields (at 14:00 LT) on the typhoon-induced and no-typhoon O₃
396 pollution days of two seasons. Large standard-exceedance (> 160 µg/m³) areas were distributed in the PRD on most days, and
397 the typhoon-induced days of both seasons generally featured higher O₃ levels. The distinct wind fields for these scenarios,
398 which were consistent with those in the longer timespan (Fig. S5), indeed led to different spatial distributions of O₃. Generally,
399 the most severe O₃ pollution occurred in the downwind areas, such as the central and southern parts of the PRD on the typhoon-
400 induced days in October 2015, the central PRD on the no-typhoon days in October 2015, and the northern and eastern PRD on
401 the no-typhoon days in July 2016. On the typhoon-induced days in July 2016, high levels of O₃ accumulated around the PRE.
402 In this section, we discuss the different contributions of various O₃ processes and sources on these days to better understand
403 the effect of typhoons on O₃ pollution in the PRD.
404

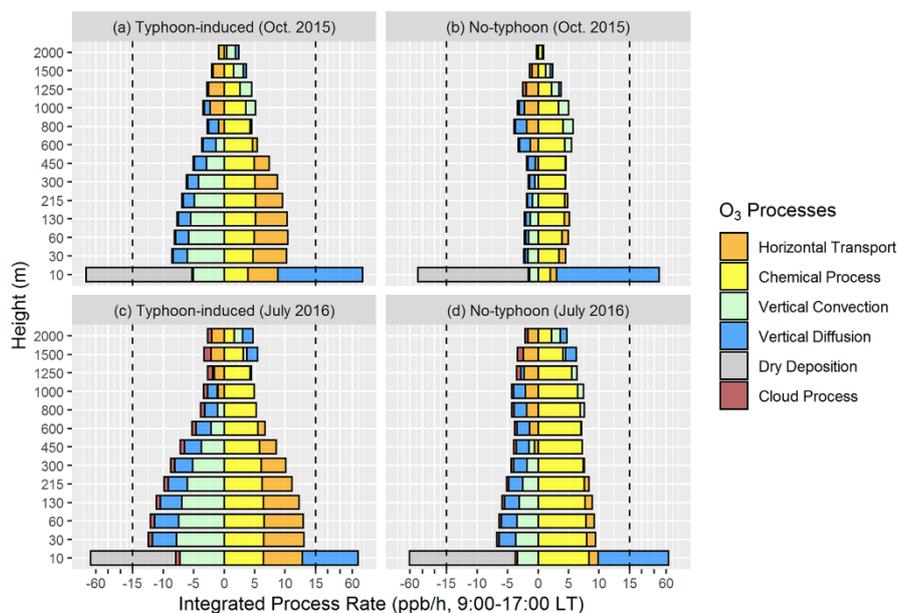


405

406 **Figure 8.** Modelling mean O₃ MDA8 concentrations (µg/m³) and wind vectors (at 14:00 LT) on the representative O₃ pollution days: (a)
407 the typhoon-induced days in October 2015 (14–16 and 21 October 2015); (b) the no-typhoon days in October 2015 (28 October and 3–5
408 November 2015); (c) the typhoon-induced days in July 2016 (7–8 and 30–31 July 2016); and (d) the no-typhoon days in July 2016 (22–26
409 and 29 July 2016). Three representative sites in the PRD are shown as black circles in the plots: XJ, Xijiao; MDS, Modiesha; DF,
410 Duanfen.

411 4.1 O₃ processes: transport vs chemical process

412 The PA tool in CMAQ was used to quantify the contributions of transport and chemical process to the O₃ variations on O₃
 413 pollution days in October 2015 and July 2016. As is shown in Fig. 9, the daytime (9:00–17:00 LT) O₃ PA results within the
 414 PRD in all scenarios share similar characteristics. Dry deposition dominated O₃ removal near the surface, and it also led to
 415 high gradients of O₃ concentrations that promote downward O₃ diffusion. Within the PBL (about 0–1 km in height), O₃ was
 416 mainly contributed by horizontal transport and chemical process, and vertical convection led to the drop of O₃ concentrations.
 417 However, differences existed between the O₃ PA results in the typhoon-induced and no-typhoon scenarios, indicating the
 418 impact of typhoons on the transport and production of O₃. In both months, typhoons led to notably higher contribution of
 419 horizontal transport to O₃, especially in the lower and middle part of the PBL. Within the PBL, on average, it increased from
 420 -0.9 ppb/h, -0.8 ppb/h to 1.2 ppb/h, 2.0 ppb/h under typhoon influence in autumn and summer, respectively. The comparison
 421 of the contribution of chemical process (in absolute rates) suggests that they had opposite effects in the two months — under
 422 typhoons, the contribution increased in October 2015 (from 4.0 ppb/h to 4.5 ppb/h within the PBL, or by 11.4%), but it
 423 decreased in July 2016 (from 7.1 ppb/h to 5.7 ppb/h within the PBL, or by -20.8%). In other words, typhoons promoted and
 424 hindered O₃ production in autumn and summer, respectively. These results agree well with the comparisons of O₃ transport
 425 and production conditions in the previous section.
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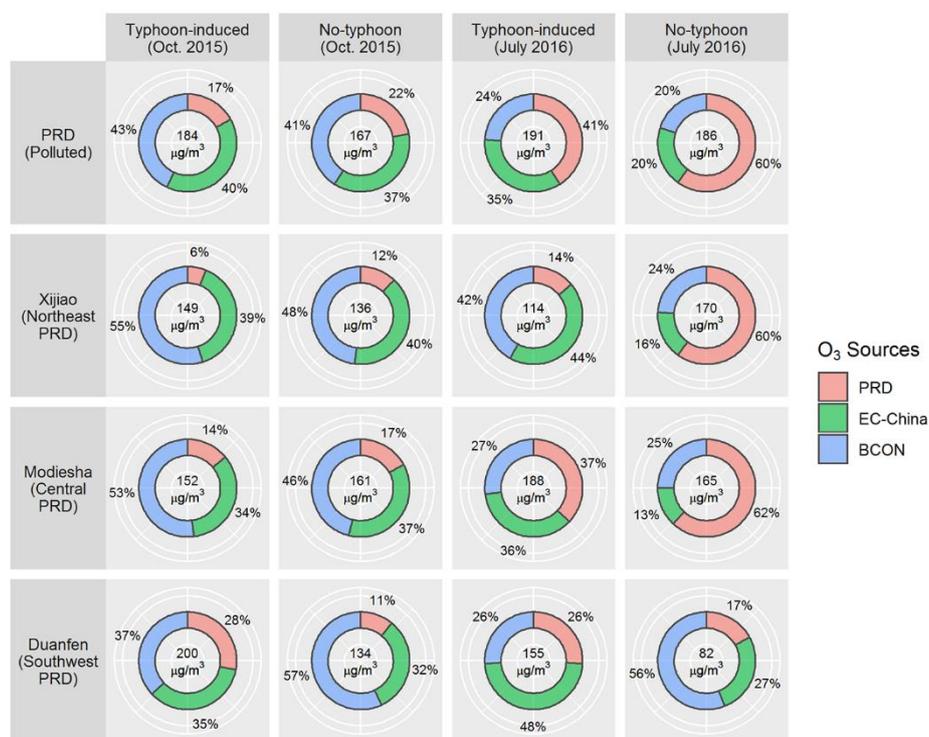


427

428 **Figure 9.** The daytime-mean (9:00–17:00 LT) hourly contributions of O₃ processes within the PRD in vertical layers 1–13 on representative
 429 O₃ pollution days: (a) the typhoon-induced days in October 2015 (14–16 and 21 October 2015); (b) the no-typhoon days in October 2015
 430 (28 October and 3–5 November 2015); (c) the typhoon-induced days in July 2016 (7–8 and 30–31 July 2016); and (d) the no-typhoon days
 431 in July 2016 (22–26 and 29 July 2016).

432 4.2 O₃ sources: local sources vs regional sources

433 The contributions of various sources to O₃ within the PRD are determined by the combined impact of O₃ transport, production
 434 and accumulation. The results for mean daytime (9:00–17:00 LT) O₃ SA near the ground (about 0–80 m in height) on typhoon-
 435 induced and no-typhoon O₃ pollution days are illustrated in Fig. 10. For polluted regions within the PRD, stronger O₃
 436 production under typhoons did not lead to a higher proportion of local contributions to O₃ pollution in October 2015 — it even
 437 decreased from 22% (on the no-typhoon days) to 17% (on the typhoon-induced days). The contributions of EC-China
 438 emissions and BCON, in contrast, increased slightly from 37%, 41% to 40%, 43%, respectively. The distinction of the O₃ SA
 439 results is more apparent for the summer scenarios, that is, typhoons resulted in growing contributions from O₃ transported from
 440 other regions (from 40% to 59%) but decreased local contributions (from 60% to 41%) in July 2016. More favourable O₃
 441 accumulation conditions (indicated by higher APRTs on the representative typhoon-induced O₃ pollution days in summer (Fig.
 442 S9)) were far from sufficient to compensate for the effect of weakened O₃ production on the high contributions of local sources.
 443



444

445 **Figure 10.** The mean O₃ SA near the ground (about 0–80 m in height) on the represented typhoon-induced and no-typhoon O₃ pollution
 446 days in October 2015 and July 2016 (the average results of 9:00–17:00 LT). The locations of the three representative sites (Xijiao,
 447 Modiesha and Duanfen) are shown in Fig. 8. PRD, the Pearl River Delta; EC-China, East China and Central China; BCON, the boundary
 448 conditions of the d02 modelling.

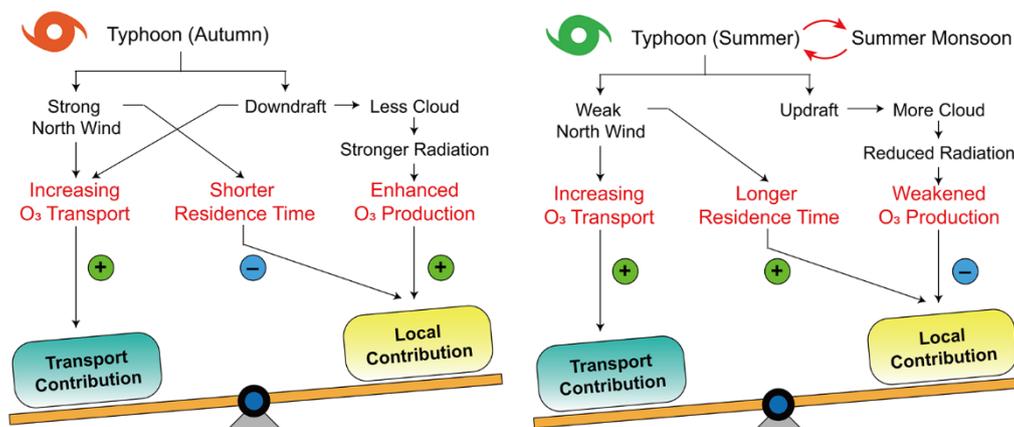
449 Furthermore, owing to the variations of wind fields, the comparison results of O₃ SA in different parts of the PRD may differ
 450 from the regional ones. For instance, while the comparisons of O₃ SA in the Xijiao and Modiesha site (located in the northeast

451 and central part of the PRD, respectively) agree well with those in the polluted regions of the PRD, higher contributions of
452 PRD emissions for O₃ can be found in the Duanfen site (located in the southwest part of the PRD) on the typhoon-induced
453 days of two months in comparison to these on the corresponding no-typhoon days (Fig. 10). Since the site was located in the
454 downwind region in the typhoon-induced scenario in October 2015 (Fig. 8a), enhanced O₃ production led by typhoons from
455 the massive emissions of O₃ precursors in the central PRD (Zheng et al., 2009) contributed to higher local contributions for O₃
456 pollution here (as the distribution of local contributions in percentage to daytime O₃ shown in Fig. S10, the highest local
457 contribution in the PRD occurred in areas near the Duanfen site and almost reached 40% in this scenario, which was even
458 higher than that in the corresponding no-typhoon scenario (33%)). In the no-typhoon scenario in July 2016, the site was located
459 in the upwind regions under the prevailing of ~~southwest~~southerly-winds, limiting the contributions of local emissions for O₃
460 at the site (Fig. 8d). Thus, higher local contributions can also be found in the typhoon-induced scenario in this month.

461 **5 Discussion and conclusions**

462 The significance of typhoons on O₃ pollution in the PRD calls for thorough evaluations of the different causes of O₃ pollution
463 with the appearance of typhoons in the Northwest Pacific. In this study, we revealed the different impacts of typhoons on O₃
464 transport, production and accumulation in the PRD (as summarised in Fig. 11) through systematic comparisons of
465 meteorological conditions, the contributions of various O₃ processes and sources in the typhoon-induced and no-typhoon
466 scenarios. We found that typhoons tended to promote O₃ transport towards the PRD, but failed to provide more favourable O₃
467 production and accumulation conditions simultaneously, which limited the contribution of local emissions to O₃ pollution.
468 Furthermore, there were also differences between the influence of typhoons on O₃ pollution in autumn and summer. More
469 favourable transport conditions occurred in the typhoon-induced scenario in autumn, which was characterised by higher wind
470 speeds and the increased influence of downdrafts. In summer, the mixed types of air masses in the typhoon-induced scenario
471 were likely to bring more O₃ into the PRD than the clean marine air masses in the no-typhoon scenario, also suggesting
472 enhanced O₃ transport under the influence of typhoons. Generally, typhoons led to cloudless conditions, stronger solar radiation,
473 and thus more rapid O₃ production in autumn, but shorter APRTs (5–10 hours) suggest that locally sourced O₃ was hard to
474 accumulate within the PRD. As a result, the contributions in percentage of local emissions to O₃ pollution decreased (slightly
475 by ~5% for the polluted regions of the PRD in October 2015). In contrast, in summer, intensified updrafts associated with
476 typhoons strengthened cloud formation, weakened solar radiation, and thus restrained local O₃ production. Longer APRTs (>
477 20 hour) under typhoon influence were far from sufficient to maintain high contributions of local emissions for O₃ pollution
478 (which decreased by ~20% for the polluted regions of the PRD in July 2016). However, due to the variations of wind fields
479 under different scenarios, the changes of local and transport contributions for O₃ led by typhoons were different in the
480 southwest part of the PRD, that is, higher contribution from emissions within the PRD and reduced transport contribution
481 occurred in the typhoon-induced scenarios in both seasons. As for the close typhoon-induced scenario, O₃ transport was further

482 strengthened, but meteorological conditions in the PRD became less favourable for both the production and accumulation of
483 O₃.
484



485
486 **Figure 11.** The summary of the causes of O₃ pollution in the PRD under typhoon influence in autumn and summer.

487 The East Asian monsoon, changing with seasons, has a pronounced impact on local meteorological conditions as well as the
488 characteristics of O₃ pollution in East China (He et al., 2008). The seasonal behaviour of the East Asian monsoon is likely to
489 result in the seasonally varied effect of typhoons on O₃ pollution in the PRD. In October, the summer monsoon has almost
490 finished its retraction and the winter monsoon is beginning (Ding, 1994). Thus, there are not many obstacles to the southward
491 movement of typhoon periphery and the transport of O₃ towards the PRD by the continental and coastal air masses. Large
492 downdraft-influenced areas in Central and South China occur in this scenario, and high O₃ levels and low RH in these areas
493 indicate the potentially important role of directly downward O₃ transport. In July, the summer monsoon reaches its strongest
494 (Ding, 1994). The interaction between typhoon periphery and the summer monsoon results in stagnation and enhanced updrafts
495 above the land areas of the PRD and its surroundings. Only when typhoon is close enough to the PRD is the stagnation
496 terminated and the structure of the summer monsoon cell broken. This also explains why some summertime typhoon-induced
497 O₃ episodes in the PRD can be typically divided into two periods, as stagnation leads to the accumulation of locally produced
498 O₃ in the first phase and strong northerly winds strengthen O₃ transport before the landing of typhoons (Lam et al., 2005; Li,
499 2013). It should be noted that updrafts, rather than downdrafts, prevailed on the typhoon-induced O₃ pollution days in summer.
500 High levels of O₃ did not necessarily occur in the regions dominated by downdrafts in this scenario, suggesting a less notable
501 connection between downdrafts and summertime O₃ pollution in the PRD. Further investigations are required to trace the
502 detailed process of downward O₃ transport, including the stratosphere-troposphere exchange (Stohl et al., 2003), in each
503 scenario, and quantify their contributions to near-ground O₃ pollution.
504

505 Some limitations remain in this study. We chose O₃ pollution days as individual samples, ignoring the influence of O₃ pollution
506 on the previous days. Thus, more detailed full-episode analyses are required. Moreover, owing to the small sampling size, the
507 influence of typhoons on O₃ pollution in the PRD is still not fully understood, including, for instance, the detailed connections
508 between the features of typhoons (intensity, position) and O₃ pollution. However, the comparisons of meteorological conditions,
509 O₃ processes and sources in different scenarios and seasons demonstrate the complex causes of typhoon-induced O₃ pollution
510 in the PRD — typhoons tend to enhance O₃ transport into the PRD in both seasons, but their impacts on the production and
511 accumulation of O₃ are completely different. As a result, emissions within (outside of) the PRD are likely to contribute less
512 (more) on the typhoon-induced O₃ pollution days than on the no-typhoon days. In order to effectively alleviate O₃ pollution
513 and to reduce the population exposure in the PRD, more attention should be paid to controlling anthropogenic emissions of O₃
514 precursors on a larger scale, rather than focusing on local emission, under typhoon influence. For air quality management, it
515 is suggested to comprehensively evaluate the ~~efficiency-response~~ of O₃ levels in the PRD to fractional local and non-local
516 ~~emission reductions to reduce O₃ levels so as to find the most effective strategies to alleviate O₃ pollution in the PRD~~ in
517 different scenarios (Thunis et al., 2019; Thunis et al., 2020). This study also suggests that a thorough evaluation of O₃ transport,
518 production and accumulation conditions can be applied to understand the causes of regional O₃ pollution not only in the PRD,
519 but also in other regions. The results will help find efficient strategies to alleviate regional O₃ pollution as well as to reduce its
520 adverse effects.

521
522 *Data availability.* Data are available from the corresponding author upon request.

523
524 *Author contributions.* KQ, XW and YZ designed the study. KQ, XW, and TX did the simulation work, including the operation
525 of the WRF, SMOKE and CMAQ models. JS, HD, LZ and YZ provided observational results of field campaigns and the
526 routine monitoring datasets for the evaluation of model performance. KQ, XW, YY and YZ analysed the modelling results.
527 KQ, XW, YY and YZ wrote and revised this paper, with critical feedbacks from all other authors.

528
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530
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