A Comparative Study to Reveal the Influence of Typhoons on the

Transport, Production and Accumulation of O₃ in the Pearl River
 Dolto, China

3 Delta, China

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14 Abstract. The Pearl River Delta (PRD) region in South China is faced with severe ambient O_3 pollution in autumn and summer, 15 which mostly coincides with the occurrence of typhoons above the Northwest Pacific. With increasingly severe O_3 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 O_3 transport, production and accumulation for efficient O_3 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 O_3 transport conditions in the typhoon-induced scenario in both seasons. 23 However, the effect of typhoons on the production and accumulation of O_3 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 O_3 within the PRD. In contrast, in summer, typhoons increased cloud cover, and 26 weakened solar radiation, thus restraining O_3 formation, but the growing residence time of local air masses favoured O_3 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 O_3 pollution, 29 confirming enhanced O_3 transport under typhoon influence in both seasons. The results of the process analysis in CMAO 30 suggest that the chemical process contributed more in autumn but less in summer in the PRD. Since O_3 production and 31 accumulation cannot be enhanced at the same time, the proportion of O_3 contributed by emissions within the PRD was likely 32 to decrease in both seasons. The difference in the typhoon influence on O_3 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

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

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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 O_3 levels in recent years (Li et al., 2019), O_3 has become the primary contributor to the deterioration of air quality in this 47 region (Feng et al., 2019). The occurrence of O_3 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 O_3 episodes (regional-mean maximum 8-h average O_3 concentrations > 240 50 μ g/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 O_3 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 O_3 pollution is closely linked to meteorological conditions that are conducive to the transport, production and/or 57 accumulation of O_3 . Stagnation caused by typhoons, characterised by low wind speeds, has been reported during many 58 episodes, and it promotes the accumulation of locally formed O_3 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). Strong north or west winds were observed or simulated during several episodes, suggesting the potentially strengthened 60 transport of pollutants under typhoon influence (Wang et al., 2001; Yang et al., 2012; Wang et al., 2015; Wei et al., 2016). 61 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

chemical transport models, along with the Process Analysis (PA) tool and source apportionment (SA) methods, to quantify 66 67 and compare the contributions of various O_3 processes (e.g., transport and the chemical process) and sources (e.g., local emissions, outside emissions and background) during these episodes. Based on reports by Huang et al. (2005), Lam et al. 68 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), horizontal/vertical transport and chemical production may both be the main contributing process for typhoon-induced O_3 70 71 pollution in different parts of the PRD. The SA results revealed that emissions within the PRD contributed 40-80% of O_3 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 O_3 transport, production and accumulation within the PRD all enhanced at the same time by typhoons? Do both 75 O_3 pollution seasons (autumn and summer) experience similar impact of typhoons on O_3 pollution? More thorough 76 investigations are needed to answer these questions.

77

In this study, we present systematic comparisons between O_3 pollution in the typhoon-induced and no-typhoon scenarios 78 79 (definitions given in Sect. 2.2) to elucidate the influence of typhoons on O_3 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 O_3 pollution days using the Community Multiscale Air Ouality (CMAQ) model, were used in the comparisons. A detailed introduction of these datasets is presented 83 in Sect. 2. The comparisons were conducted from the perspectives of meteorological conditions (Sect. 3), O₃ processes and 84 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:

- Re-analysis datasets: We mainly used the ERA-Interim re-analysis product in the comparisons due to its 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 analyses data, including air temperature (at a height of 2 m),
- 95 relative humidity (RH, at 1000 hPa), horizontal wind speeds (at a height of 10 m; zonal and meridional wind speeds
- 96 were also involved in the comparisons), and low (for the height at which pressure/surface pressure > 0.8), medium (for
- 97 the height at which 0.45 < pressure/surface pressure < 0.8), high (for the height at which pressure/surface pressure <

- 98 0.45) and total cloud covers; (2) near-surface parameters from the forecast data, including plenary boundary layer
- 99 (PBL) height and net surface solar radiation; and (3) upper air parameters at multiple heights, including horizontal and
- 100 vertical wind speeds, cloud water content and O₃ mixing ratio. The focus of this study is O₃ pollution during the
- daytime, and therefore, only the parameters at 14:00 local time (LT) were selected for the analyses (except for net
 surface solar radiation, which was averaged within 8:00–17:00 LT).
- Surface meteorological routine monitoring datasets: The routine monitoring meteorological data collected at 29
 national meteorological sites within the PRD (locations shown in Fig. S1a) were also used to explore the
 meteorological features under the impact of typhoons. The parameters include air temperature, RH, and wind speed and
 direction (also transformed to zonal and meridional wind speeds in the comparisons) at 14:00 LT.
- Typhoon information: The times, locations and intensities of typhoons were provided by the Chinese Meteorological
 Administration Best Track Dataset of tropical cyclones (Ying et al., 2014). The tracks of all typhoons that potentially
 contributed to O₃ pollution in the PRD during the study period (October and July in 2014–2018) are shown in Fig. S2
 and S3.
- 111 **O**₃ 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
- 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**

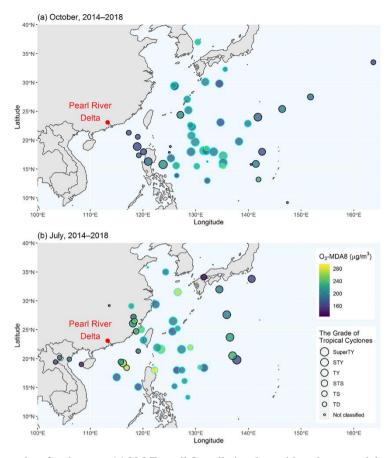
117 In this study, O_3 pollution days were defined as the days when the MDA1 exceeds 200 μ g/m³ or the MDA8 exceeds 160 118 μ g/m³ for O₃ (both are the Grade-II thresholds of the Chinese National Ambient Air Quality Standard (NAAQS), GB 3095-119 2012) in any of the nine municipalities in the PRD. According 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 $\sim 30\%$ days of both months. Typhoons tend to result in more severe O₃ pollution in the PRD, as 125 indicated by generally higher O_3 MDA1 and MDA8 values with the influence of typhoons than these without typhoons.

- 126
- 127 The differing locations of typhoons can result in the diverse effect of typhoons on O₃ pollution (Chow et al., 2018). To
- 128 determine the general influence of typhoons on O₃ pollution in the PRD, it was necessary to further select O₃ pollution days
- coinciding with typhoons with similar directions and distances to the PRD. As is shown in Fig. 1, all O₃ pollution days in
- 130 October and most O_3 pollution days in July under typhoon influence were associated with typhoons to the east of the PRD,

131 which were more likely to cause O_3 pollution (Chow et al., 2018). In order to minimize the disturbance of typhoon directions 132 in the comparisons, we removed the remaining five O₃ pollution days in July with typhoons located to the due north or 133 southwest of the PRD from the analyses. After this, based on the distances between typhoon centres and the PRD (at 14:00 134 LT), we classified the pollution days with typhoons in each season into three categories: close typhoon (lowest 20% of 135 distances), typhoon (20–80% intervals of distances), and far typhoon (longest 20% of distances)-induced days. The typhoon-136 induced days represent O₃ pollution days with general typhoon influence, and they were compared with those without the 137 appearance of typhoons (hereafter denoted as the no-typhoon days). It should be noted that the distances between typhoon 138 centres and the PRD on the typhoon-induced days were overall larger in autumn (1400-2800 km, at 14:00 LT) than in 139 summer (700–2000 km, at 14:00 LT), which may be the consequence of the different characteristics of typhoon paths in the 140 two seasons: most typhoons in autumn travel northwest initially and then turn northward in the areas east of the Philippines 141 (Fig. S2), whereas they are more likely to end up landing in Southeast China in summer (Fig. S3). Since the influence of 142 typhoons on O_3 pollution may be different when typhoons come close enough to the PRD (Lam et al., 2005; Li, 2013), the 143 close typhoon-induced days were considered to be a special scenario in the comparisons of meteorological conditions (Sect. 3.5). Owing to the less apparent effect of typhoons over the PRD, we did not include the far typhoon-induced days in the 144 145 discussions.

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14/	Table I. The	numbers and	proportions	OI U3 1	pollution c	lays,	and O_3	concentrations	for various	scenarios.

Parameter	October, 2014–2018	July, 2014–2018 1	148	
Number (proportion) of O3 pollution days	78 (50.3%)	55 (35.5%) 1	149	
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		



150

Figure 1. The location and intensity of typhoons at 14:00 LT on all O₃ pollution days with typhoons, and the corresponding O₃ MDA8
 concentrations (maximum values in the nine municipalities of the PRD) on the same days during (a) October and (b) July in 2014–2018.
 The points with cyan borders indicate the "typhoon-induced" O₃ pollution days used in the comparisons. The grades of tropical cyclones
 (Chinese National Standard, GB/T 19201-2006) are as follows: SuperTY - super typhoon; STY - severe typhoon; TY - typhoon; STS -

155 severe tropical storm; TS - tropical storm; TD - tropical depression; others are grouped as "not classified".

156 2.3 Calculation of the trajectories and air parcel residence time

157 To explore the potential effect of cross-regional transport on O₃ pollution in the PRD, we applied the Hysplit model (Stein et

- 158 al., 2015) with the Global Data Assimilation System (GDAS) datasets as inputs to calculate 72-h backward trajectories reaching
- 159 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
- 160 part of the PRD, was the endpoint of backward trajectories. Its height was set to 500 m above the ground to indicate the effect
- 161 of long-range transport on O₃ pollution within the PBL (Park et al., 2007).
- 162
- 163 Air parcel residence time (APRT), discussed by Huang et al. (2019), is the average number of hours that air parcels originated
- 164 from one place stay within a pre-defined domain, and long APRTs can be used to indicate good accumulation conditions for
- 165 locally sourced pollutants. To calculate APRTs in the PRD, we designed a 21×15 point matrix (resolution: 0.2 °×0.2 °) that

embraces the whole PRD (Fig. S4), and forward trajectories starting from these points were also calculated using the Hysplit 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 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. Afterwards, the length of time each trajectory remained within the administration borders of the PRD, i.e., APRT, was calculated and attributed to its starting point. APRTs in each point were averaged, and these averaged APRT values in all points were interpolated using the Kriging method to obtain field results for the further comparisons.

172 2.4 CMAQ modelling: basic setups and modelling methods

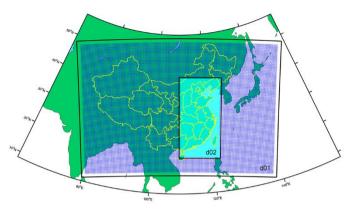
173 We utilised the widely used 3D chemical transport model CMAQ (version 5.0.2) to investigate the effects of typhoons on O_3 174 processes and sources. October 2015 and July 2016 featured the most severe O₃ pollution under typhoon influence among all 175 representative months in autumn and summer, respectively (Table S3). Therefore, O_3 pollution in these two months were 176 modelled and used in further comparisons. Note that the modelling period in autumn was adjusted to 11 October-10 November 177 2015 to involve more O_3 pollution days (3–5 November 2015). In detail, there were four typhoon-induced O_3 pollution days 178 (14–16 and 21 October 2015) and four no-typhoon O₃ pollution days (28 October and 3–5 November 2015) in October 2015, 179 whereas there were four and six typhoon-induced and no-typhoon days in July 2016, respectively (typhoon-induced: 7–8 and 180 30-31 July 2016; and no-typhoon: 22–26 and 29 July 2016). The results of daytime (9:00–17:00 LT) O₃ PA and SA on the 181 above O_3 pollution days were averaged for the typhoon-induced and no-typhoon scenarios in autumn (October 2015) and 182 summer (July 2016) and were used in the comparisons.

183

184 The main setups of the CMAQ model are presented as follows. Two-nested modelling domains with the resolutions of 36 and 185 12 km (denoted as d01 and d02, respectively) were set in this study (Fig. 2). Specifically, d02 covers the whole East and 186 Central China (EC-China), enabling us to evaluate the contribution of emissions in these areas to O_3 pollution in the PRD. 187 There were 19 vertical layers in the CMAQ modelling, with about 10 layers within the PBL (about 0-1 km in heights; Guo et 188 al., 2016). The Weather Research and Forecasting (WRF) model (version 3.2) provided the meteorological fields used as inputs. 189 SMOKE (version 2.5) and MEGAN (version 2.10) were used to process the anthropogenic and biogenic emissions, 190 respectively. The anthropogenic emission inventory used in this study consisted of the following three parts: (1) emissions in 191 the PRD, which were provided by the Guangdong Environmental Monitoring Centre; (2) emissions in other areas of mainland 192 China, which were extracted from the MEIC inventory (He, 2012); and (3) emissions in other countries and regions in Asia, 193 which were extracted from the MIX inventory (Li et al., 2017). The initial and boundary conditions of the d01 modelling were 194 obtained from the same-period results of the MOZART-4 global model (available at https://www.acom.ucar.edu/wrf-195 chem/mozart.shtml, last accessed: Dec. 2019), and those of the d02 modelling were extracted from the d01 modelling results. 196 The SAPRC07 gas-phase chemistry mechanism (Carter, 2010) and the AERO6 aerosol scheme were set in the modelling. In 197 addition, the simulations of the two months were both started 10 days ahead to minimise the disturbance of the bias of the 198 initial conditions. The modelling performances of CMAQ and WRF were determined to be acceptable based on the

comparisons between the observational and modelling series of meteorological parameters, O_3 MDA8, daily NO_2 concentrations and the mixing ratios of non-methane hydrocarbons (NMHCs) in the PRD (for details, refer to Sect. 1 in the Supplement Information), which ensures the validity of the further analyses.

202



203

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

The PA tool in CMAQ was implemented to quantify the hourly contributions of O_3 processes (or integrated process rate, IPR), which includes vertical/horizontal transport (convection+diffusion), chemical process (net O_3 production through gas-phase reactions), dry deposition and cloud process. To explore the overall effect of typhoons on O_3 transport and production in the region, the mean PA results within the administration boundaries of the PRD were calculated and compared.

210

211 In order to identify the sources of all O₃ in the PRD, we used the classic Brute Force Method (BFM) to identify the contributions 212 of emissions (including anthropogenic and biogenic emissions) in the PRD and other regions in the d02 (mainly EC-China), 213 as well as regions outside the d02 (the boundary conditions of the d02) for O_3 pollution in the PRD (hereafter denoted as the contributions of PRD, EC-China and BCON, or S_{PRD}, S_{EC-China} and S_{BCON}, respectively). For a pollutant, the contribution of a 214 215 specific emission, E_i, can be calculated in two ways: (1) the difference between the modelled concentrations of the base case 216 (all emissions involved) and the sensitivity case where E_i is zeroed out (i.e., top-down BFM); (2) the difference between two 217 sensitivity cases where emissions except E_i and all of the emissions are zeroed out, respectively (i.e., bottom-up BFM). Owing 218 to the non-linearity between O₃ and its precursors, biases may occur between the results of two types of BFM methods, leading 219 to the non-additivity of the results (Clappier et al., 2017). Therefore, the average of the top-down and bottom-up BFM results 220 was treated as the quantified contributions of the concerned sources. Four simulation cases were run in this study, including 221 (the modelled O₃ concentration in each case was also marked in brackets):

- 222 the base case (C_{base}) ;
- the PRD-cut case ($C_{PRD_{cut}}$), where emissions within the PRD were zeroed out;
- the PRD-only case (C_{PRD_only}), where emissions outside the PRD (within d02) were zeroed out; and

- the zero-emission case (C_0) , where all emissions within the d02 were zeroed out.
- 226 Afterwards, the S_{PRD}, S_{EC-China} and S_{BCON} values (in concentrations) in the polluted areas of the PRD (where modelled daytime
- $227 \qquad O_3 \ concentrations > 160 \ \mu g/m^3, the \ Grade-II \ O_3 \ MDA8 \ thresholds \ of the \ Chinese \ NAAQS) \ were \ calculated \ using the following \ MDA8 \ thresholds \ of the \ Chinese \ NAAQS) \ were \ calculated \ using \ the \ following \ MDA8 \ thresholds \ of \ the \ Chinese \ NAAQS) \ were \ calculated \ using \ the \ following \ MDA8 \ thresholds \ of \ the \ Chinese \ NAAQS) \ were \ calculated \ using \ the \ following \ the \ following \ MDA8 \ thresholds \ of \ the \ Chinese \ NAAQS) \ were \ calculated \ using \ the \ following \ MDA8 \ thresholds \ of \ the \ Chinese \ NAAQS) \ were \ calculated \ using \ the \ following \ thresholds \$

228 equations,

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

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

$$S_{BCON} = C_0. \tag{R3}$$

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

230 3 Comparison of meteorological conditions

231 3.1 Overview: comparison of meteorological parameters in the PRD

232 First, we compared near-ground meteorological parameters in the PRD on the typhoon-induced and no-typhoon O₃ pollution 233 days. Near-surface parameters from routine monitoring datasets and the ERA-Interim re-analysis were used in the 234 comparison. The parameters include air temperature, RH, wind speeds, cloud covers, PBL height and net surface solar 235 radiation. Since there was no rainfall on most O₃ pollution days (indicated by the weather in Guangzhou (Table S4)), 236 precipitation was not considered in the comparisons. For consistency, the parameters of ERA-Interim were extracted at the 237 same time (14:00 LT) and the locations of sites (Fig. S1a) as those in routine monitoring. The Mann-Whitney U test was 238 applied to determine whether the above parameters were significantly different (p < 0.05) between typhoon-induced and no-239 typhoon O_3 pollution days.

240

241 As is listed in Table 2, statistically significant differences between the typhoon-induced and no-typhoon scenarios existed for 242 most of the parameters, such as meridional (south-north) wind speed, cloud covers within various height ranges and net 243 surface solar radiation — in both seasons, these parameters were significantly different for the two scenarios. It indicates that 244 the causes of O₃ pollution may vary on typhoon-induced and no-typhoon O₃ pollution days. Note that air temperature, one of 245 the parameters most closely related to O_3 pollution in the PRD (Zhao et al., 2019), was not significantly different in the two 246 scenarios. We also found that the comparison in autumn and summer did not produce the same results: the typhoon-induced days in autumn featured lower RH, stronger winds (especially north wind), reduced cloud cover (low, medium, high and 247 248 total) and stronger surface solar radiation, whereas in summer, these days had higher RH, weaker south winds, more cloud 249 cover (medium, high and total), weaker surface solar radiation and lower PBL heights. Therefore, the impact of typhoons on 250 O_3 pollution differs in the two seasons, as well. In order to reveal the impact of typhoons on O_3 transport, production, and 251 accumulation in the PRD, more detailed comparisons of the corresponding meteorological indicators are presented in the 252 following sections.

253

254 Table 2. The comparisons of meteorological parameters (all at 14:00 LT except for net surface solar radiation, which is the average value

255 for 9:00–17:00 LT) in the PRD for the three scenarios (no-typhoon, typhoon-induced, close typhoon-induced) in two seasons (autumn,

summer). RM, routine measurement; ERA, ERA-Interim re-analysis. All of the parameters are presented as "the mean value ± standard

257 deviation". The differences between parameters in the typhoon-induced or close typhoon-induced scenarios and the corresponding

258 typhoon-induced scenarios for the same season are given in parentheses, and "*" indicates p < 0.05, or statistically significant differences

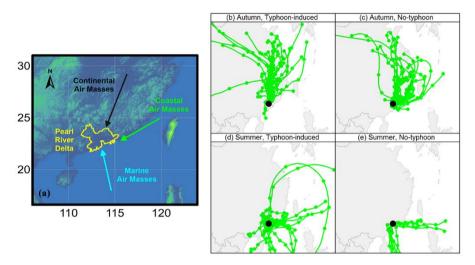
	D .	Autumn (October, 2014–2018)			Summer (July, 2014–2018)			
Parameters	Data Source	No-typhoon	Typhoon-induced	Close Typhoon- induced	No-typhoon	Typhoon-induced	Close Typhoon- induced	
Air Temperature	RM	29.1 ± 2.2	29.3 ± 1.8	29.6 ± 1.5	33.7 ± 2.0	33.9 ± 2.0	35.0 ± 1.5	
(°C)			(+0.2)	(+0.5, *)		(+0.2)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

260 3.2 O3 transport conditions: comparison of wind speeds, backward trajectories and vertical air motions

261 The higher wind speeds and/or O_3 levels in the transported air masses are, the more likely O_3 transport plays an increasingly

262 important role in O₃ pollution. In the PRD, O₃ levels are closely linked to the type of air masses influencing the region, which

263 can be identified based on backward trajectories. According to Zheng et al. (2010), there are generally three types of air masses 264 that are transported into the PRD along different paths and contribute to O_3 pollution here, namely, the continental, coastal and 265 marine air masses (Fig. 3a). The continental and coastal air masses can bring O_3 from EC-China to the PRD, and thus, they 266 are typically recognised as being polluted and contributing to relatively high O_3 levels in the PRD. In contrast, the marine air 267 masses, originated from the South China Sea, are much cleaner. In this section, we studied the influence of typhoons on O_3 268 transport by comparing wind speeds and 72-h backward trajectories in the typhoon-induced and no-typhoon scenarios.



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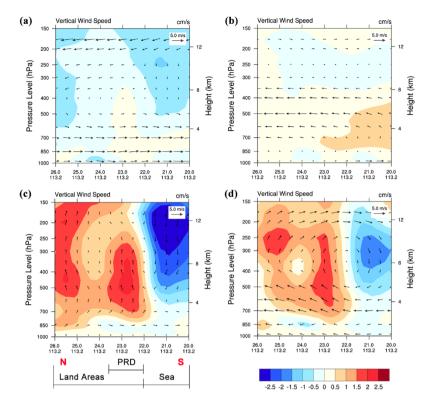
271Figure 3. (a) Three O_3 transport paths towards the PRD. (b–e) Backward trajectories at 14:00 LT for each scenario: (b) autumn, typhoon-272induced; (c) autumn, no-typhoon; (d) summer, typhoon-induced; and (e) summer, no-typhoon. The black dots indicate the end point of all273trajectories, i.e., where the Modiesha site in the central PRD is located.

274 As is displayed in Fig. 3b–c, we identified the influence of continental air masses on the typhoon-induced O_3 pollution days 275 in autumn, as well as mixed contributions from the continental and coastal air masses on the corresponding no-typhoon days. 276 However, for the former scenario, significantly increased wind speeds (Table 2) ensure more favourable conditions for the 277 transport of O_3 . In summer, the three types of air masses may all have affected O_3 pollution in the typhoon-induced scenario, 278 while only the marine air masses influenced the PRD in the no-typhoon scenario (Fig. 3d-e). The increasing influence of much 279 more polluted air masses (continental and coastal air masses) led by typhoon ensured that more O_3 was transported to the PRD, 280 thus typhoons also tended to increase the contribution of transport to O_3 pollution in the PRD in summer. In addition, the 281 influence of different air masses was also accompanied with variations in the prevailing winds in the PRD, that is, north winds 282 and easterlies in the typhoon-induced and no-typhoon scenarios in autumn, respectively, and southerly winds in the no-typhoon 283 scenario in summer (indicated by wind roses in Fig. S5). For the typhoon-induced scenario in summer, the dominate wind 284 direction is hard to determine. These variations in the local wind fields potentially result in the different spatial distribution of 285 O₃ concentrations in various scenarios.

287 Downdrafts are typically considered to be an important cause of typhoon-induced O₃ pollution (Lam, 2018), but in which 288 scenarios downdrafts influence the PRD remains unclear. Thus, we explored the overall features of vertical air motions from 289 the surface layer to the tropopause in the typhoon-induced and no-typhoon scenarios, and the ERA-Interim reanalysis dataset 290 (including all upper air parameters at multiple heights introduced in Sect. 2.1) was utilised in the comparisons. The contours 291 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-292 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 293 (Fig. S4). On the typhoon-induced days in autumn, downdrafts occurred over large areas above the PRD, especially above a 294 height of ~700 hPa. Although updrafts can still be found near the sea surface in this scenario, vertical wind speeds tended to 295 be lower compared with those on the no-typhoon days in autumn, which also suggests the enhancement of downdrafts caused 296 by typhoons. In summer, the influence of downdrafts was found over the PRD under 850 hPa on the typhoon-induced O_3 297 pollution days. However, overall, updrafts prevailed above the land areas and downdrafts prevailed above the sea in both the 298 typhoon-induced and no-typhoon scenarios in summer, which is recognised as the structure of the East Asian summer monsoon 299 cell (Chen et al., 1964; Jin et al., 2013; Ding et al., 2018). For both updrafts and downdrafts, the absolute values of vertical wind speeds in the typhoon-induced scenario in summer were overall higher than those in the corresponding no-typhoon 300 301 scenario. Therefore, the approach of typhoons did not break the structure of the summer monsoon cell, but rather they further 302 strengthened the vertical motions above both land areas and sea. These analyses suggest that typhoons do not necessarily lead 303 to downdrafts during O_3 pollution periods in the PRD and its adjacent areas; and in summer, vertical air motions affected by 304 typhoons are more complicated than expected owing to the existence of the East Asian summer monsoon.

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306 We also explored the regions where downdrafts and updrafts occurred on a larger scale and their potential connections with 307 O₃ levels. As is shown in Fig. 5, though updrafts appeared in the PRD at 850 hPa on the typhoon-induced days in autumn, 308 downdrafts dominated in the region at 700 and 500 hPa. For the areas to the north of the PRD, the important role of downdrafts 309 was found at all three heights. In contrast to the no-typhoon days in autumn, downdrafts tended to cover much larger areas in 310 this scenario. Moreover, these areas at 850 and 700 hPa generally featured higher O₃ mixing ratios as well as lower RH (Fig. 311 S6) than others, which is a sign of possible direct downward O_3 transport (Roux et al., 2020; Wang et al., 2020). This part of 312 O_3 can notably aggravate near-ground O_3 pollution in the PRD. In contrast, in summer, updrafts dominated the PRD at various 313 heights in both scenarios. Besides the PRD, most of the regions near the coast were characterised by updrafts above the land 314 as well as downdrafts offshore, further indicating the ubiquity of the summer monsoon cell. By comparing the two scenarios in summer, we found that typhoons resulted in more areas being influenced by updrafts. The areas with high O₃ levels did not 315 316 coincide with the downdraft-affected areas, and therefore, O_3 transported from the upper air may play a less significant role in 317 the typhoon-induced O_3 pollution in summer.



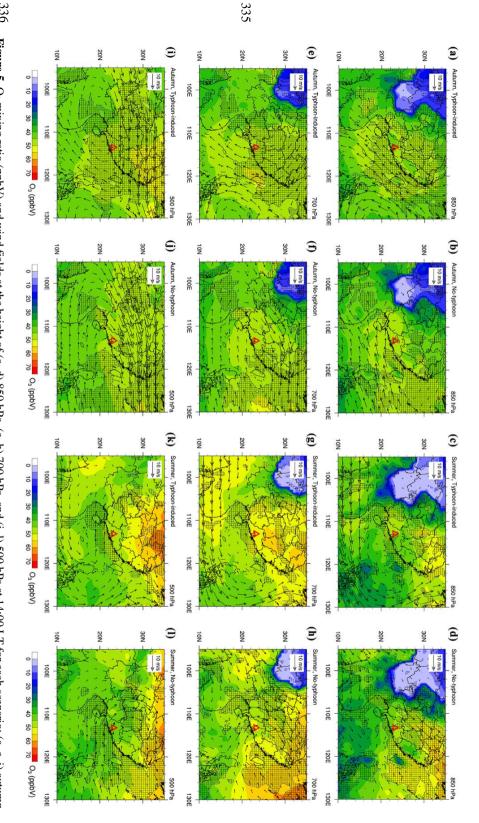
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Figure 4. The cross sections of mean vertical wind field at 14:00 LT for each scenario: (a) autumn, typhoon-induced; (b) autumn, notyphoon; (c) summer, typhoon-induced; and (d) summer, no-typhoon. Cross sections were made along the 113.2 E longitude 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), and the contours indicate

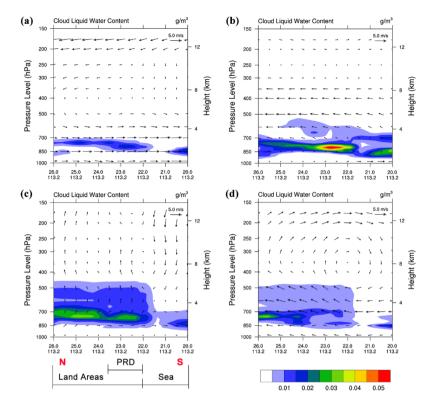
322 vertical wind speed (cm/s). PRD, the Pearl River Delta.

323 **3.3** O₃ production conditions: comparison of clouds

324 Clouds efficiently reflect solar radiation (Liou, 1976), and therefore, they have a notable impact on the local formation of O_3 . 325 Figure 6 displays the cross sections of mean ERA-Interim cloud liquid water contents (CLWC) at 14:00 LT of all O₃ pollution 326 days corresponding to the typhoon-induced and no-typhoon scenarios of two seasons, which were also made along the 113.2 °E 327 longitude line, from 26.0 N to 20.0 N (Fig. S4). The comparison of CLWC in the cross sections suggests that typhoons 328 generally resulted in fewer clouds in autumn but more clouds in summer, which agrees well with the comparison of cloud 329 covers in Table 2. The presence of fewer clouds on the typhoon-induced days in autumn can be attributed to two reasons: the 330 influence of dry air masses (indicated by lower RH in Table 2 and Fig. S6) and/or the hindrance of cloud formation by 331 downdrafts. In summer, the strengthened updrafts above the land caused by typhoons favoured cloud formation, which is 332 demonstrated by higher CLWC at the heights of 500-850 hPa and increases in medium and high cloud covers. In areas above 333 the PRD below 850 hPa, downdrafts led to slight decrease of clouds in the typhoon-induced scenario in summer, which is also 334 indicated by reduced low cloud cover. As a consequence of varied cloud covers in each scenario, on average, net surface solar



336 337 338 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, 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 PRD. The gridded areas indicate that vertical wind speed is less than 0, or downdrafts occur.



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Figure 6. The cross sections of mean cloud liquid water content (g/m³) and wind vectors at 14:00 LT for each scenario: (a) autumn, typhooninduced; (b) autumn, no-typhoon; (c) summer, typhoon-induced; and (d) summer, no-typhoon. Cross sections were made along the 113.2 E 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 Pearl River Delta.

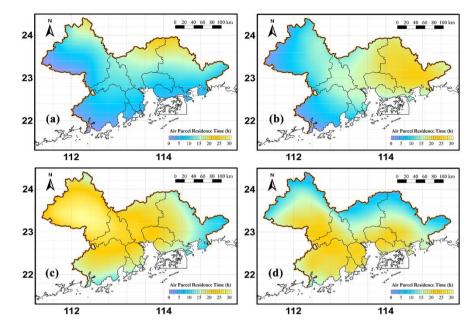
radiation increased by 13% and decreased by 7% on the typhoon-induced days in autumn and summer, respectively (Table 2),

345 which promoted and hindered O₃ production in the PRD during these two seasons, respectively.

346 3.4 O₃ accumulation conditions: comparison of APRTs

347 The longer APRTs are, the more likely that O_3 produced by local emissions accumulates within the targeted region and notably 348 contributes to near-ground O₃ pollution. In order to study the effect of typhoons on O₃ accumulation, we calculated APRTs in 349 the PRD in the typhoon-induced and no-typhoon scenarios (Fig. 7) for the further comparisons. On the typhoon-induced days 350 in autumn, APRTs were typically 5-10 hours (mean = 9.5 hours) — shorter than those on the no-typhoon days in autumn 351 (mean = 13.1 hours). In addition, lower APRT values occurred in the central part of the PRD, where high anthropogenic 352 emissions of pollutants are distributed (Zheng et al., 2009). Despite more active O₃ chemistry discussed in the last section, 353 locally sourced O₃ was less likely to accumulate within the PRD in this scenario, potentially limiting the contribution of local 354 emissions for O₃. The comparison suggests opposite results in the summer scenarios, that is, APRTs on the typhoon-induced 355 days (20–30 hours, mean = 21.0 hours) were overall higher than those on the no-typhoon days (15–25 hours, mean = 16.5356 hours). This favoured the accumulation of locally sourced O_3 . Based on the comparisons in this and previous section, typhoons

- 357 did not provide more favourable conditions for O₃ production and accumulation simultaneously in the PRD in both autumn
- and summer, thus potentially resulting in reduced contributions in percentage of local emissions to O_3 pollution here. More quantitative evaluations of the contributions from multiple O_3 sources are discussed in Sect. 4.
- 360



361

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

364 3.5 Meteorological conditions on the close typhoon-induced days

365 On the close typhoon-induced days in the two seasons, stronger north winds prevailed and total cloud cover was higher than 366 that on the no-typhoon days (Table 2), suggesting better conditions for the transport of O_3 but less favourable conditions for 367 O₃ production. As displayed in Fig. S7, the APRT values were significantly lower on the close typhoon-induced days (mean = 6.6 hours, 12.9 hours in autumn and summer, respectively) than on the no-typhoon days, making it even harder for locally 368 369 sourced O_3 to accumulate within the PRD. Therefore, close typhoons are concluded to promote the transport of O_3 from the 370 outside and to reduce the contributions of O₃ produced from local emissions in a more notable way. In addition, close typhoons 371 led to stronger downdrafts in autumn and updrafts in summer than other scenarios in the same season (Fig. S8). It should be 372 noted that the structure of the summer monsoon cell near the PRD was destroyed in the close typhoon-induced scenario in 373 summer, indicating the stronger influence of typhoons on regional wind fields. The dominant role of O_3 transport during O_3 374 pollution days in this special scenario agrees well with the reported episode-based analyses (Lam et al., 2005; Li, 2013).

375 4 Comparisons of O₃ processes and sources

376 The comparisons of meteorological conditions served as qualitative evidence to determine the general influence of typhoons 377 on O₃ transport, production and accumulation in autumn and summer. Based on the comparison between the CMAO modelling 378 results on typical O₃ pollution days in October 2015 and July 2016, more quantitative evidence can be presented. Figure 8 379 displays modelled mean O₃ MDA8 concentrations and wind fields (at 14:00 LT) on the typhoon-induced and no-typhoon O₃ 380 pollution days of two seasons. Large standard-exceedance (> $160 \mu g/m^3$) areas were distributed in the PRD on most days, and 381 the typhoon-induced days of both seasons generally featured higher O_3 levels. The distinct wind fields for these scenarios, 382 which were consistent with those in the longer timespan (Fig. S5), indeed led to different spatial distributions of O₃. Generally, 383 the most severe O₃ pollution occurred in the downwind areas, such as the central and southern parts of the PRD on the typhoon-384 induced days in October 2015, the central PRD on the no-typhoon days in October 2015, and the northern and eastern PRD on 385 the no-typhoon days in July 2016. On the typhoon-induced days in July 2016, high levels of O_3 accumulated around the PRE. 386 In this section, we discuss the different contributions of various O₃ processes and sources on these days to better understand 387 the effect of typhoons on O₃ pollution in the PRD.

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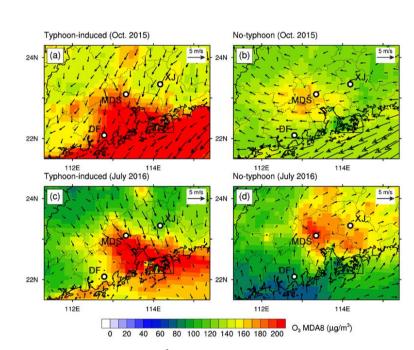


Figure 8. Modelling mean O_3 MDA8 concentrations ($\mu g/m^3$) and wind vectors (at 14:00 LT) on the representative O_3 pollution days: (a)

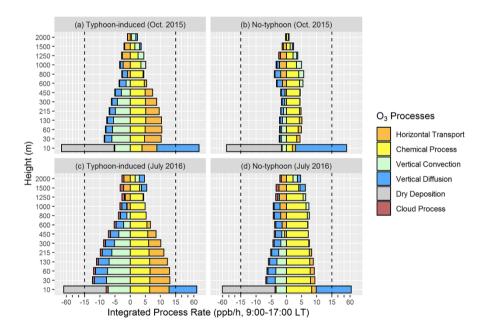
- 393 and 29 July 2016). Three representative sites in the PRD are shown as black circles in the plots: XJ, Xijiao; MDS, Modiesha; DF,
- 394 Duanfen.

<sup>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
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</sup>

395 4.1 O₃ processes: transport vs chemical process

396 The PA tool in CMAO was used to quantify the contributions of transport and chemical process to the O_3 variations on O_3 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 397 398 PRD in all scenarios share similar characteristics. Dry deposition dominated O₃ removal near the surface, and it also led to 399 high gradients of O_3 concentrations that promote downward O_3 diffusion. Within the PBL (about 0–1 km in height), O_3 was 400 mainly contributed by horizontal transport and chemical process, and vertical convection led to the drop of O_3 concentrations. However, differences existed between the O₃ PA results in the typhoon-induced and no-typhoon scenarios, indicating the 401 402 impact of typhoons on the transport and production of O_3 . In both months, typhoons led to notably higher contribution of 403 horizontal transport to O₃, especially in the lower and middle part of the PBL. Within the PBL, on average, it increased from 404 -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 405 of the contribution of chemical process (in absolute rates) suggests that they had opposite effects in the two months — under 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 406 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 407 408 hindered O_3 production in autumn and summer, respectively. These results agree well with the comparisons of O_3 transport 409 and production conditions in the previous section.

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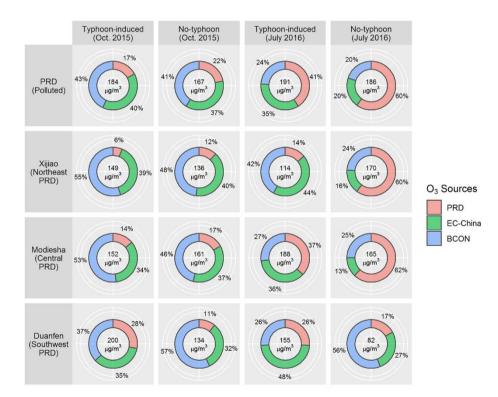


412 Figure 9. The daytime-mean (9:00–17:00 LT) hourly contributions of O₃ processes within the PRD in vertical layers 1–13 on representative

- 413 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
- 414 (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
- 415 in July 2016 (22–26 and 29 July 2016).

416 4.2 O₃ sources: local sources vs regional sources

417 The contributions of various sources to O_3 within the PRD are determined by the combined impact of O_3 transport, production 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-418 419 induced and no-typhoon O_3 pollution days are illustrated in Fig. 10. For polluted regions within the PRD, stronger O_3 420 production under typhoons did not lead to a higher proportion of local contributions to O_3 pollution in October 2015 — it even 421 decreased from 22% (on the no-typhoon days) to 17% (on the typhoon-induced days). The contributions of EC-China 422 emissions and BCON, in contrast, increased slightly from 37%, 41% to 40%, 43%, respectively. The distinction of the O₃ SA 423 results is more apparent for the summer scenarios, that is, typhoons resulted in growing contributions from O₃ transported from 424 other regions (from 40% to 59%) but decreased local contributions (from 60% to 41%) in July 2016. More favourable O_3 425 accumulation conditions (indicated by higher APRTs on the representative typhoon-induced O₃ pollution days in summer (Fig. 426 (S9) were far from sufficient to compensate for the effect of weakened O_3 production on the high contributions of local sources. 427



428

- 429 **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
- 430 days in October 2015 and July 2016 (the average results of 9:00–17:00 LT). The locations of the three representative sites (Xijiao,
- Modiesha and Duanfen) are shown in Fig. 8. PRD, the Pearl River Delta; EC-China, East China and Central China; BCON, the boundary
 conditions of the d02 modelling.

433 Furthermore, owing to the variations of wind fields, the comparison results of O₃ SA in different parts of the PRD may differ

434 from the regional ones. For instance, while the comparisons of O₃ SA in the Xijiao and Modiesha site (located in the northeast

435 and central part of the PRD, respectively) agree well with those in the polluted regions of the PRD, higher contributions of 436 PRD emissions for O_3 can be found in the Duanfen site (located in the southwest part of the PRD) on the typhoon-induced 437 days of two months in comparison to these on the corresponding no-typhoon days (Fig. 10). Since the site was located in the 438 downwind region in the typhoon-induced scenario in October 2015 (Fig. 8a), enhanced O₃ production led by typhoons from 439 the massive emissions of O_3 precursors in the central PRD (Zheng et al., 2009) contributed to higher local contributions for O_3 440 pollution here (as the distribution of local contributions in percentage to daytime O_3 shown in Fig. S10, the highest local 441 contribution in the PRD occurred in areas near the Duanfen site and almost reached 40% in this scenario, which was even 442 higher than that in the corresponding no-typhoon scenario (33%)). In the no-typhoon scenario in July 2016, the site was located in the upwind regions under the prevailing of southerly winds, limiting the contributions of local emissions for O_3 at the site 443 444 (Fig. 8d). Thus, higher local contributions can also be found in the typhoon-induced scenario in this month.

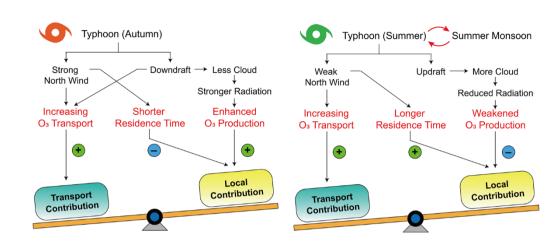
445 **5 Discussion and conclusions**

446 The significance of typhoons on O_3 pollution in the PRD calls for thorough evaluations of the different causes of O_3 pollution 447 with the appearance of typhoons in the Northwest Pacific. In this study, we revealed the different impacts of typhoons on O_3 448 transport, production and accumulation in the PRD (as summarised in Fig. 11) through systematic comparisons of 449 meteorological conditions, the contributions of various O_3 processes and sources in the typhoon-induced and no-typhoon 450 scenarios. We found that typhoons tended to promote O_3 transport towards the PRD, but failed to provide more favourable O_3 451 production and accumulation conditions simultaneously, which limited the contribution of local emissions to O_3 pollution. 452 Furthermore, there were also differences between the influence of typhoons on O_3 pollution in autumn and summer. More 453 favourable transport conditions occurred in the typhoon-induced scenario in autumn, which was characterised by higher wind 454 speeds and the increased influence of downdrafts. In summer, the mixed types of air masses in the typhoon-induced scenario 455 were likely to bring more O_3 into the PRD than the clean marine air masses in the no-typhoon scenario, also suggesting enhanced O_3 transport under the influence of typhoons. Generally, typhoons led to cloudless conditions, stronger solar radiation, 456 457 and thus more rapid O_3 production in autumn, but shorter APRTs (5–10 hours) suggest that locally sourced O_3 was hard to 458 accumulate within the PRD. As a result, the contributions in percentage of local emissions to O_3 pollution decreased (slightly 459 by ~5% for the polluted regions of the PRD in October 2015). In contrast, in summer, intensified updrafts associated with 460 typhoons strengthened cloud formation, weakened solar radiation, and thus restrained local O_3 production. Longer APRTs (> 20 hour) under typhoon influence were far from sufficient to maintain high contributions of local emissions for O_3 pollution 461 (which decreased by ~20% for the polluted regions of the PRD in July 2016). However, due to the variations of wind fields 462 under different scenarios, the changes of local and transport contributions for O_3 led by typhoons were different in the 463 464 southwest part of the PRD, that is, higher contribution from emissions within the PRD and reduced transport contribution 465 occurred in the typhoon-induced scenarios in both seasons. As for the close typhoon-induced scenario, O₃ transport was further

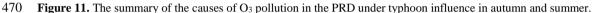
466 strengthened, but meteorological conditions in the PRD became less favourable for both the production and accumulation of

467 O₃.





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471 The East Asian monsoon, changing with seasons, has a pronounced impact on local meteorological conditions as well as the 472 characteristics of O_3 pollution in East China (He et al., 2008). The seasonal behaviour of the East Asian monsoon is likely to 473 result in the seasonally varied effect of typhoons on O_3 pollution in the PRD. In October, the summer monsoon has almost 474 finished its retraction and the winter monsoon is beginning (Ding, 1994). Thus, there are not many obstacles to the southward 475 movement of typhoon periphery and the transport of O_3 towards the PRD by the continental and coastal air masses. Large 476 downdraft-influenced areas in Central and South China occur in this scenario, and high O₃ levels and low RH in these areas 477 indicate the potentially important role of directly downward O_3 transport. In July, the summer monsoon reaches its strongest 478 (Ding, 1994). The interaction between typhoon periphery and the summer monsoon results in stagnation and enhanced updrafts 479 above the land areas of the PRD and its surroundings. Only when typhoon is close enough to the PRD is the stagnation 480 terminated and the structure of the summer monsoon cell broken. This also explains why some summertime typhoon-induced 481 O₃ episodes in the PRD can be typically divided into two periods, as stagnation leads to the accumulation of locally produced 482 O₃ in the first phase and strong northerly winds strengthen O₃ transport before the landing of typhoons (Lam et al., 2005; Li, 483 2013). It should be noted that updrafts, rather than downdrafts, prevailed on the typhoon-induced O_3 pollution days in summer. 484 High levels of O_3 did not necessarily occur in the regions dominated by downdrafts in this scenario, suggesting a less notable 485 connection between downdrafts and summertime O_3 pollution in the PRD. Further investigations are required to trace the 486 detailed process of downward O_3 transport, including the stratosphere-troposphere exchange (Stohl et al., 2003), in each 487 scenario, and quantify their contributions to near-ground O₃ pollution.

489 Some limitations remain in this study. We chose O₃ pollution days as individual samples, ignoring the influence of O₃ pollution 490 on the previous days. Thus, more detailed full-episode analyses are required. Moreover, owing to the small sampling size, the 491 influence of typhoons on O_3 pollution in the PRD is still not fully understood, including, for instance, the detailed connections 492 between the features of typhoons (intensity, position) and O₃ pollution. However, the comparisons of meteorological conditions, 493 O_3 processes and sources in different scenarios and seasons demonstrate the complex causes of typhoon-induced O_3 pollution 494 in the PRD — typhoons tend to enhance O₃ transport into the PRD in both seasons, but their impacts on the production and 495 accumulation of O_3 are completely different. As a result, emissions within (outside of) the PRD are likely to contribute less 496 (more) on the typhoon-induced O_3 pollution days than on the no-typhoon days. In order to effectively alleviate O_3 pollution and to reduce the population exposure in the PRD, more attention should be paid to controlling anthropogenic emissions of O_3 497 498 precursors on a larger scale, rather than focusing on local emission, under typhoon influence. For air quality management, it 499 is suggested to comprehensively evaluate the response of O₃ levels in the PRD to fractional local and non-local emission 500 reductions so as to find the most effective strategies to alleviate O_3 pollution in different scenarios (Thunis et al., 2019; Thunis 501 et al., 2020). This study also suggests that a thorough evaluation of O_3 transport, production and accumulation conditions can 502 be applied to understand the causes of regional O_3 pollution not only in the PRD, but also in other regions. The results will 503 help find efficient strategies to alleviate regional O_3 pollution as well as to reduce its adverse effects.

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505 Data availability. Data are available from the corresponding author upon request.

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507 Author contributions. KQ, XW and YZ designed the study. KQ, XW, and TX did the simulation work, including the operation 508 of the WRF, SMOKE and CMAQ models. JS, HD, LZ and YZ provided observational results of field campaigns and the 509 routine monitoring datasets for the evaluation of model performance. KQ, XW, YY and YZ analysed the modelling results. 510 KQ, XW, YY and YZ wrote and revised this paper, with critical feedbacks from all other authors.

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512 Competing interests. The authors declare no conflict of interest.

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