# A Comparative Study to Reveal the Influence of Typhoons on the

Transport, Production and Accumulation of O<sub>3</sub> 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 O<sub>3</sub> pollution in the PRD via systematic comparisons of meteorological conditions, O<sub>3</sub> processes and sources on 19 O<sub>3</sub> 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 O<sub>3</sub> 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 O<sub>3</sub> 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 O<sub>3</sub> pollution in the PRD and their seasonal differences. To alleviate O<sub>3</sub> pollution under

35 typhoon influence, emission control is needed on a larger scale, rather than only within the PRD.

# 36 1 Introduction

Tropospheric ozone (O<sub>3</sub>) 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 O<sub>3</sub> is produced from its precursors, i.e., nitrogen oxides (NO<sub>x</sub> = NO + NO<sub>2</sub>) and volatile organic compounds (VOCs), through chemical reactions in the presence of sunlight. Due to the relatively long lifetime of O<sub>3</sub> (~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 O<sub>3</sub> can all contribute to the O<sub>3</sub> 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 O<sub>3</sub> 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  $\mu$ g/m<sup>3</sup>) 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<sub>3</sub> 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 O<sub>3</sub> has

53 important implications for efficient and strategic O<sub>3</sub> reduction in the PRD.

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55 Analyses of typhoon-related O<sub>3</sub> 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 O<sub>3</sub> transport and contribute to near-ground O<sub>3</sub> 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 O<sub>3</sub> production, which is another important cause of O<sub>3</sub> 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), 70 horizontal/vertical transport and chemical production may both be the main contributing process for typhoon-induced  $O_3$ pollution in different parts of the PRD. The SA results revealed that emissions within the PRD contributed 40–80% of  $O_3$ 71 during typhoon-related O<sub>3</sub> episodes (Li et al., 2012; Li, 2013; Chen et al., 2015), suggesting the potentially important role of 72 73 O<sub>3</sub> accumulation for O<sub>3</sub> 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), O<sub>3</sub> processes and 84 85 sources (Sect. 4), and the conclusions about the influence of typhoons on the causes of ambient O<sub>3</sub> 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 due to its more available 91 parameters and high spatial coverage (available at https://www.ecmwf.int/en/forecasts/datasets/reanalysis-datasets/era-92 interim, last accessed: March 2020; Dee et al., 2011; Berrisford et al., 2011). Specifically, meteorological parameters 93 used in the comparisons include the following three categories: (1) near-surface parameters from the analysis fields, 94 including air temperature (at a height of 2 m), relative humidity (RH, at 1000 hPa), horizontal wind speeds (at a height 95 of 10 m; zonal and meridional wind speeds were also involved in the comparisons), and low (for the height at which 96 pressure/surface pressure > 0.8), medium (for the height at which 0.45 < pressure/surface pressure < 0.8), high (for the 97 height at which pressure/surface pressure < 0.45) and total cloud covers; (2) near-surface parameters from the forecast

- 98 fields, including plenary boundary layer (PBL) height and net surface solar radiation; and (3) upper air parameters at
- 99 multiple heights, including horizontal and vertical wind speeds, cloud water content and O<sub>3</sub> mixing ratio. The focus of
- 100 this study is  $O_3$  pollution during the daytime, and therefore, only the parameters at 14:00 local time (LT) were selected 101 for the analyses (except for net surface solar radiation, which was averaged within 8:00–17:00 LT).
- 102 ٠ 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
- 104 meteorological features under the impact of typhoons. The parameters include air temperature, RH, and wind speed and 105 direction (also transformed to zonal and meridional wind speeds in the comparisons) at 14:00 LT.
- 106 **Typhoon information:** The times, locations and intensities of typhoons were provided by the Chinese Meteorological ٠ 107 Administration Best Track Dataset of tropical cyclones (Ying et al., 2014). The tracks of all typhoons that potentially 108 contributed to  $O_3$  pollution in the PRD during the study period (October and July in 2014–2018) are shown in Fig. S2 109 and S3.
- 110  $O_3$  concentrations: Hourly  $O_3$  concentration data, which were originally released by the China National Environmental
- 111 Monitoring Centre, were downloaded from http://beijingair.sinaapp.com (last accessed: Dec. 2018). Based on the
- 112 hourly data, we calculated the maximum 1-hr concentrations (MDA1) and maximum 8-hr average concentrations
- 113 (MDA8) of O<sub>3</sub> in nine municipalities in the PRD (including Guangzhou, Shenzhen, Zhuhai, Foshan, Jiangmen,
- 114 Zhaoqing, Huizhou, Dongguan and Zhongshan) to identify  $O_3$  pollution days that served as samples in the comparisons.

#### 2.2 Definition and classification of O<sub>3</sub> pollution days 115

116 In this study,  $O_3$  pollution days were defined as the days when the MDA1 exceeds 200  $\mu$ g/m<sup>3</sup> or the MDA8 exceeds 160 117  $\mu$ g/m<sup>3</sup> for O<sub>3</sub> (both are the Grade-II thresholds of the Chinese National Ambient Air Quality Standard (NAAQS), GB 3095-118 2012) in any of the nine municipalities in the PRD. According to these criteria, there were 78 and 55  $O_3$  pollution days 119 (given in Table S1 and S2) during October and July in 2014–2018, respectively. The information about these O<sub>3</sub> pollution 120 days in the two representative months is listed in Table 1 (overall) and S3 (monthly), including the numbers of days, their 121 proportions in the month, and the corresponding mean O<sub>3</sub> concentrations (MDA8 and MDA1, highest values among nine 122 municipalities in the PRD). Although there were more O<sub>3</sub> pollution days in October than in July, O<sub>3</sub> pollution under typhoon 123 influence occurred on  $\sim$  30% days of both months. Higher O<sub>3</sub> MDA1 and MDA8 values can be found with the appearance of 124 typhoons in comparison with days without typhoons in July, whereas these values are similar in October, indicating the 125 important role of typhoons in O<sub>3</sub> pollution in the PRD.

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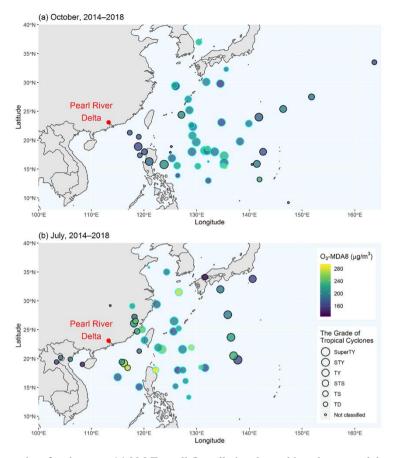
- 127 The differing locations of typhoons can result in the diverse effect of typhoons on  $O_3$  pollution (Chow et al., 2018). To
- 128 determine the general influence of typhoons on  $O_3$  pollution in the PRD, it was necessary to further select  $O_3$  pollution days
- 129 coinciding with typhoons with similar directions and distances to the PRD. As is shown in Fig. 1, all O<sub>3</sub> pollution days in
- 130 October and most O<sub>3</sub> 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 O<sub>3</sub> 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 O<sub>3</sub> 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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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 <sub>3</sub> MDA8 (µg/m <sup>3</sup> )				
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 <sub>3</sub> MDA1 (µg/m <sup>3</sup> )				
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

151 Figure 1. The location and intensity of typhoons at 14:00 LT on all O<sub>3</sub> pollution days with typhoons, and the corresponding O<sub>3</sub> MDA8 152 concentrations (maximum values in the nine municipalities of the PRD) on the same days during (a) October and (b) July in 2014–2018. 153 The points with cyan borders indicate the "typhoon-induced" O<sub>3</sub> 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 severe tropical storm; TS - tropical storm; TD - tropical depression; others are grouped as "not classified".

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# 156 **2.3 Calculation of the trajectories and air parcel residence time**

157 To explore the potential effect of cross-regional transport on O<sub>3</sub> 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 O<sub>3</sub> 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 as 500 m above the ground to better represent

161 the effect of long-range transport on O<sub>3</sub> pollution, as well as to minimize the disturbance of objects near the surface to the

162 transport (Park et al., 2007).

163

164 Air parcel residence time (APRT), discussed by Huang et al. (2019), is the average number of hours that air parcels originated

165 from one place stay within a pre-defined domain, and long APRTs can be used to indicate good accumulation conditions for

locally sourced pollutants. To calculate APRTs in the PRD, we designed a  $21 \times 15$  point matrix (resolution:  $0.2 \times 0.2$ ) 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 O<sub>3</sub> 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.

# 173 2.4 CMAQ modelling: basic setups and modelling methods

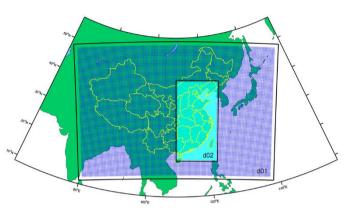
174 We utilised the widely used 3D chemical transport model CMAQ (version 5.0.2) to investigate the effects of typhoons on  $O_3$ 175 processes and sources. October 2015 and July 2016 featured the most severe O<sub>3</sub> pollution under typhoon influence among all 176 representative months of the two seasons (Table S3), and thus, they were chosen as the period in the modelling (because there 177 was no severe  $O_3$  pollution during the first 10 days of October 2015 and 3–5 November can be classified as the no-typhoon  $O_3$ 178 pollution days, we adjusted the modelling period in autumn to 11 October-10 November 2015) and all typhoon-induced and 179 no-typhoon  $O_3$  pollution days in these two months served as representative  $O_3$  pollution days in the comparisons. In detail, 180 there were four typhoon-induced O<sub>3</sub> pollution days (14–16 and 21 October 2015) and four no-typhoon O<sub>3</sub> pollution days (28 181 October and 3–5 November 2015) in October 2015, whereas there were four and six typhoon-induced and no-typhoon days in 182 July 2016, respectively (typhoon-induced: 7–8 and 30–31 July 2016; and no-typhoon: 22–26 and 29 July 2016). The results of 183 daytime (9:00-17:00 LT) O<sub>3</sub> PA and SA on the above O<sub>3</sub> pollution days were averaged for the typhoon-induced and no-184 typhoon scenarios in autumn (October 2015) and summer (July 2016) and were used in the comparisons.

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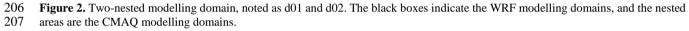
186 The main setups of the CMAQ model are presented as follows. Two-nested modelling domains with the resolutions of 36 and 187 12 km (denoted as d01 and d02, respectively) were set in this study (Fig. 2). Specifically, d02 covers the whole East and 188 Central China (EC-China), enabling us to evaluate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

addition, the simulations of the two months were both started 10 days ahead to minimise the disturbance of the bias of the 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.

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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.

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In order to identify the sources of all O<sub>3</sub> in the PRD, we used the classic Brute Force Method (BFM) to identify the contributions 213 214 of emissions (including anthropogenic and biogenic emissions) in the PRD and other regions in the d02 (mainly EC-China), 215 as well as regions outside the d02 (the boundary conditions of the d02) for  $O_3$  pollution in the PRD (hereafter denoted as the 216 contributions of PRD, EC-China and BCON, or S<sub>PRD</sub>, S<sub>EC-China</sub> and S<sub>BCON</sub>, respectively). For a pollutant, the contribution of a 217 specific emission,  $E_i$ , can be calculated in two ways: (1) the difference between the modelled concentrations of the base case 218 (all emissions involved) and the sensitivity case where  $E_i$  is zeroed out (i.e., top-down BFM); (2) the difference between two 219 sensitivity cases where emissions expect  $E_i$  and all of the emissions are zeroed out, respectively (i.e., bottom-up BFM). Owing 220 to the non-linearity between O<sub>3</sub> and its precursors, biases may occur between the results of two types of BFM methods, leading 221 to the non-additivity of the results (Clappier et al., 2017). Therefore, the average of the top-down and bottom-up BFM results 222 was treated as the quantified contributions of the concerned sources. Four simulation cases were run in this study, including 223 (the modelled  $O_3$  concentration in each case was also marked in brackets):

• 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.
- Afterwards, the  $S_{PRD}$ ,  $S_{EC-China}$  and  $S_{BCON}$  values (in concentrations) in the polluted areas of the PRD (where modelled daytime O<sub>3</sub> concentrations > 160 µg/m<sup>3</sup>, the Grade-II O<sub>3</sub> MDA8 thresholds of the Chinese NAAQS) were calculated using the following equations,

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

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

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

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

# 232 3 Comparison of meteorological conditions

# 233 3.1 Overview: comparison of meteorological parameters in the PRD

234 First, we compared near-ground meteorological parameters in the PRD on the typhoon-induced and no-typhoon O<sub>3</sub> pollution 235 days. The parameters from routine monitoring datasets (including air temperature, RH, wind speed, zonal and meridional 236 wind speeds measured at 14:00 LT of all O<sub>3</sub> pollution days at 29 national meteorological sites within the PRD (Fig. S1a)) 237 and the ERA-Interim re-analysis (including all near-surface parameters from the analysis and forecast fields introduced in 238 Sect. 2.1, extracted at the same time and the locations of sites as these in routine monitoring datasets) were used in the 239 comparison (since all O<sub>3</sub> pollution days in October and over 60% of O<sub>3</sub> pollution days in July were characterized with sunny, 240 cloudy, or overcast weathers with no rainfall in the PRD (Table S4, represented by the weather in Guangzhou), precipitation 241 was not considered in the comparisons). The Mann-Whitney U test was applied to determine whether the above parameters 242 were significantly different (p < 0.05) between typhoon-induced and no-typhoon O<sub>3</sub> pollution days.

243

244 As is listed in Table 2, statistically significant differences between the typhoon-induced and no-typhoon scenarios existed for 245 most of the parameters, such as meridional (south-north) wind speed, cloud covers within various height ranges and net 246 surface solar radiation — in both seasons, these parameters were significantly different for the two scenarios. It indicates that the causes of O<sub>3</sub> pollution may vary on typhoon-induced and no-typhoon O<sub>3</sub> pollution days. Note that air temperature, one of 247 248 the parameters most closely related to  $O_3$  pollution in the PRD (Zhao et al., 2019), was not significantly different in the two 249 scenarios. We also found that the comparison in autumn and summer did not produce the same results: the typhoon-induced 250 days in autumn featured lower RH, stronger winds (especially north wind), reduced cloud cover (low, medium, high and 251 total) and stronger surface solar radiation, whereas in summer, these days had higher RH, weaker south winds, more cloud 252 cover (medium, high and total), weaker surface solar radiation and lower PBL heights. Therefore, the impact of typhoons on

253 O<sub>3</sub> pollution differs in the two seasons, as well. In order to reveal the impact of typhoons on O<sub>3</sub> transport, production, and

accumulation in the PRD, more detailed comparisons of the corresponding meteorological indicators are presented in the

255 following sections.

256

257	Table 2. The comparisons of meteorological parameters (all at 14:00 LT except for net surface solar radiation, which is the average value
258	for 9:00-17:00 LT) in the PRD for the three scenarios (no-typhoon, typhoon-induced, close typhoon-induced) in two seasons (autumn,
259	summer). RM, routine measurement; ERA, ERA-Interim re-analysis. All of the parameters are presented as "the mean value ± standard
260	deviation." The differences between assumptions in the tembers induced an electronic induced energies and the company dimensional set of the temperature of temperature of the temperature of

260 deviation". The differences between parameters in the typhoon-induced or close typhoon-induced scenarios and the corresponding 261 typhoon-induced scenarios for the same season are given in parentheses, and "\*" indicates p < 0.05, or statistically significant differences

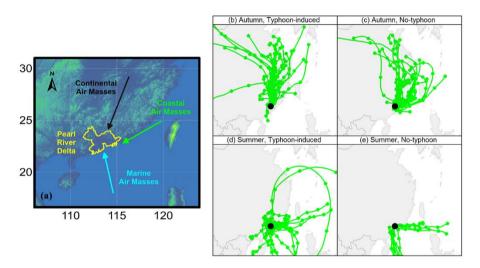
262 between these parameters when the Mann-Whitney U test is used.

	Data	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	$29.1 \pm 2.2$	$29.3 \pm 1.8$	$29.6 \pm 1.5$	$33.7 \pm 2.0$	$33.9 \pm 2.0$	$35.0 \pm 1.5$	
(°C)			(+0.2)	(+0.5, *)		(+0.2)	(+1.3, *)	
	ERA	$29.2~{\pm}2.1$	$29.3~{\pm}1.6$	$29.6 \pm 1.5$	$33.4~{\pm}1.8$	$33.5~\pm1.4$	$34.6\ \pm 1.4$	
			(+0.1)	(+0.4, *)		(+0.1)	(+1.2, *)	
RH (%)	RM	$52.4\ \pm 10.2$	$44.8~{\pm}10.4$	$51.4 \pm 12.4$	$57.0~{\pm}9.3$	$58.3~{\pm}9.7$	$56.9~{\pm}6.4$	
			(-7.6, *)	(-1.0)		(+1.3)	(-0.1)	
	ERA	$54.0 \pm 9.8$	$48.3 \pm 11.2$	$52.2 \pm 12.4$	$62.6 \pm 10.8$	$66.4 \pm 9.4$	$62.5 \pm 9.4$	
			(-5.7, *)	(-1.8, *)		(+3.8, *)	(+0.1)	
Wind Speed	RM	$2.33 \pm 1.18$	$2.58 \pm 1.23$	$2.96 \pm 1.40$	$2.46 \pm 1.33$	$2.30 \pm 1.20$	$2.53 \pm 1.16$	
(m/s)			(+0.25, *)	(+0.63, *)		(-0.16)	(+0.07)	
	ERA	$2.39~{\pm}1.30$	$2.54 \pm 0.99$	$3.53 \pm 1.11$	$2.41 \pm 0.99$	$2.18 \pm 1.18$	$2.61 \pm 1.05$	
			(+0.15, *)	(+1.14, *)		(-0.23, *)	(+0.20)	
Zonal (East-West)	RM	$-0.83 \pm 1.72$	$-0.59 \pm 1.70$	$-0.13 \pm 1.74$	$-0.41 \pm 2.05$	$-0.03 \pm 1.94$	$0.73 \pm 1.98$	
Wind Speed			(+0.24, *)	(+0.70, *)		(+0.38)	(+1.14, *)	
(m/s)	ERA	$-1.41 \pm 1.43$	$-1.07 \pm 1.04$	$-0.87 \pm 0.79$	$0.22 \pm 1.73$	$-0.02 \pm 1.81$	$0.29 \pm 2.45$	
			(+0.34, *)	(+0.54, *)		(-0.24)	(+0.07)	
Meridional (South-	RM	$-0.36 \pm 1.74$	$-1.49 \pm 1.66$	$-2.21 \pm 1.66$	$0.79 \pm 1.69$	$0.01 \pm 1.72$	$-0.69 \pm 1.68$	
North) Wind Speed			(-1.13, *)	(-1.85, *)		(-0.78, *)	(-1.48, *)	
(m/s)	ERA	$-0.27 \pm 1.82$	$-1.97 \pm 1.16$	$-3.27 \pm 1.29$	$1.61 \pm 1.09$	$0.64 \pm 1.58$	$-0.68 \pm 1.19$	
			(-1.70, *)	(-3.00, *)		(-0.97, *)	(-2.29, *)	
Low Cloud	ERA	$17.2 \pm 22.7$	$4.2 \pm 11.9$	15.5 ±23.9	$8.7 \pm 9.4$	$7.1 \pm 9.5$	$5.2 \pm 5.0$	
Cover (%)			(-13.0, *)	(-1.7, *)		(-1.6, *)	(-3.5, *)	
Medium Cloud	ERA	$22.2 \pm 26.5$	$10.4 \pm 19.7$	9.5 ±14.5	$8.7 \pm 11.1$	$15.4 \pm 15.1$	$21.5 \pm 15.5$	
Cover (%)			(-11.8, *)	(-12.7, *)		(+6.7, *)	(+12.8, *)	
High Cloud	ERA	$12.1 \pm 23.1$	$7.2 \pm 16.3$	$34.6 \pm 35.6$	$32.2 \pm 30.0$	$44.9 \pm 29.3$	$51.0 \pm 34.2$	
Cover (%)			(-4.9, *)	(+22.5, *)		(+12.7, *)	(+18.8, *)	
Total Cloud	ERA	$43.5 \pm 32.3$	$20.5 \pm 25.7$	$51.9 \pm 33.1$	$43.7 \pm 26.7$	$58.3 \pm 22.7$	$67.5 \pm 21.0$	
Cover (%)			(-23.0, *)	(+8.4, *)		(+14.6, *)	(+23.7, *)	
Net Surface Solar	ERA	$456.9 \pm 78.4$	$516.6 \pm 66.7$	$516.5 \pm 62.8$	$560.3 \pm 93.1$	$523.2 \pm 74.4$	$541.9 \pm 54.0$	
Radiation (W/m <sup>2</sup> )			(+59.7, *)	(+59.6, *)		(-37.1, *)	(-18.4, *)	
PBL Height (m)	ERA	$1471 \pm 315$	1473 ±348	1349 ±227	$1268 \pm 383$	1037 ±289	1196 ±300	
			(+2)	(-122, *)		(-231, *)	(-72, *)	

# 263 **3.2** O<sub>3</sub> transport conditions: comparison of wind speeds, backward trajectories and vertical air motions

264 The higher wind speeds and/or  $O_3$  levels in the transported air masses are, the more likely  $O_3$  transport plays an increasingly 265 important role in  $O_3$  pollution. In the PRD,  $O_3$  levels are closely linked to the type of air masses influencing the region, which 266 can be identified based on backward trajectories. According to Zheng et al. (2010), there are generally three types of air masses that are transported into the PRD along different paths and contribute to  $O_3$  pollution here, namely, the continental, coastal and 267 268 marine air masses (Fig. 3a). The continental and coastal air masses can bring  $O_3$  from EC-China to the PRD, and thus, they are typically recognised as being polluted and contributing to relatively high O<sub>3</sub> levels in the PRD. In contrast, the marine air 269 270 masses, originated from the South China Sea, are much cleaner. In this section, we studied the influence of typhoons on  $O_3$ 271 transport by comparing wind speeds and 72-h backward trajectories in the typhoon-induced and no-typhoon scenarios.

272



# 273

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

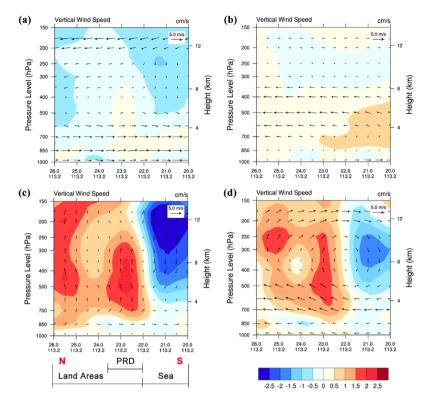
277 As is displayed in Fig. 3b–c, we identified the influence of continental air masses on the typhoon-induced  $O_3$  pollution days 278 in autumn, as well as mixed contributions from the continental and coastal air masses on the corresponding no-typhoon days. 279 However, for the former scenario, significantly increased wind speeds (Table 2) ensure more favourable conditions for the 280 transport of  $O_3$ . In summer, the three types of air masses may all have affected  $O_3$  pollution in the typhoon-induced scenario, 281 while only the marine air masses influenced the PRD in the no-typhoon scenario (Fig. 3d-e). The increasing influence of much more polluted air masses (continental and coastal air masses) led by typhoon ensured that more O<sub>3</sub> was transported to the PRD, 282 283 thus typhoons also tended to increase the contribution of transport to  $O_3$  pollution in the PRD in summer. In addition, the 284 influence of different air masses was also accompanied with variations in the prevailing winds in the PRD, that is, north winds 285 and easterlies in the typhoon-induced and no-typhoon scenarios in autumn, respectively, and southwest winds in the no-286 typhoon scenario in summer (indicated by wind roses in Fig. S5). For the typhoon-induced scenario in summer, the dominate wind direction is hard to determine. These variations in the local wind fields potentially result in the different spatial distribution of  $O_3$  concentrations in various scenarios.

289

290 Downdrafts are typically considered to be an important cause of typhoon-induced O<sub>3</sub> pollution (Lam, 2018), but in which 291 scenarios downdrafts influence the PRD remains unclear. Thus, we explored the overall features of vertical air motions from 292 the surface layer to the tropopause in the typhoon-induced and no-typhoon scenarios, and the ERA-Interim reanalysis dataset 293 (including all upper air parameters at multiple heights introduced in Sect. 2.1) was utilised in the comparisons. The contours 294 in Fig. 4 show the cross sections of mean vertical wind speed at 14:00 LT of all O<sub>3</sub> pollution days corresponding to the typhoon-295 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 296 (Fig. S4). On the typhoon-induced days in autumn, downdrafts occurred over large areas above the PRD, especially above a 297 height of ~700 hPa. Although updrafts can still be found near the sea surface in this scenario, vertical wind speeds tended to 298 be lower compared with those on the no-typhoon days in autumn, which also suggests the enhancement of downdrafts caused 299 by typhoons. In summer, the influence of downdrafts was found over the PRD under 850 hPa on the typhoon-induced  $O_3$ 300 pollution days. However, overall, updrafts prevailed above the land areas and downdrafts prevailed above the sea in both the typhoon-induced and no-typhoon scenarios in summer, which is recognised as the structure of the East Asian summer monsoon 301 302 cell (Chen et al., 1964; Jin et al., 2013; Ding et al., 2018). For both updrafts and downdrafts, the absolute values of vertical 303 wind speeds in the typhoon-induced scenario in summer were overall higher than these in the corresponding no-typhoon 304 scenario. Therefore, the approach of typhoons did not break the structure of the summer monsoon cell, but rather they further 305 strengthened the vertical motions above both land areas and sea. These analyses suggest that typhoons do not necessarily lead 306 to downdrafts during  $O_3$  pollution periods in the PRD and its adjacent areas; and in summer, vertical air motions affected by 307 typhoons are more complicated than expected owing to the existence of the East Asian summer monsoon.

308

309 We also explored the regions where downdrafts and updrafts occurred on a larger scale and their potential connections with 310 O<sub>3</sub> levels. As is shown in Fig. 5, though updrafts appeared in the PRD at 850 hPa on the typhoon-induced days in autumn, 311 downdrafts dominated in the region at 700 and 500 hPa. For the areas to the north of the PRD, the important role of downdrafts 312 was found at all three heights. In contrast to the no-typhoon days in autumn, downdrafts tended to cover much larger areas in 313 this scenario. Moreover, these areas at 850 and 700 hPa generally featured higher O<sub>3</sub> mixing ratios as well as lower RH (Fig. 314 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  $O_3$  can notably aggravate near-ground  $O_3$  pollution in the PRD. In contrast, in summer, updrafts dominated the PRD at various 315 316 heights in both scenarios. Besides the PRD, most of the regions near the coast were characterised by updrafts above the land 317 as well as downdrafts offshore, further indicating the ubiquity of the summer monsoon cell. By comparing the two scenarios 318 in summer, we found that typhoons resulted in more areas being influenced by updrafts. The areas with high  $O_3$  levels did not 319 coincide with the downdraft-affected areas, and therefore,  $O_3$  transported from the upper air may play a less significant role in 320 the typhoon-induced  $O_3$  pollution in summer.

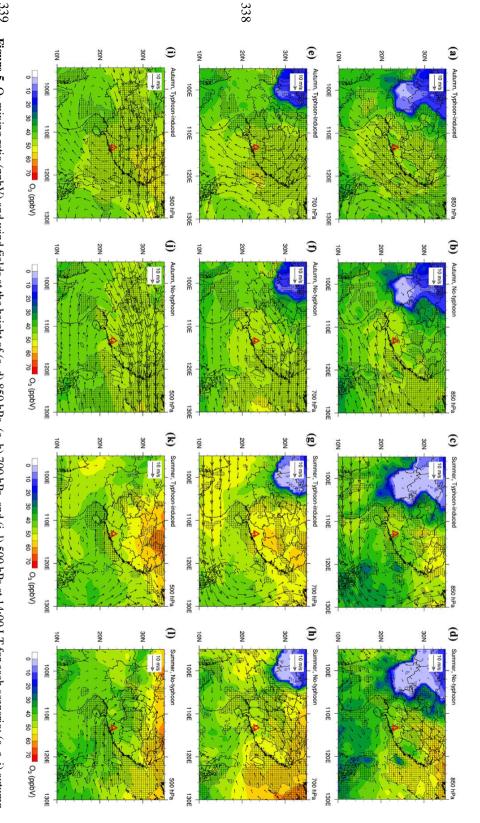


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Figure 4. The cross sections of mean vertical wind field at 14:00 LT for each scenario: (a) autumn, typhoon-induced; (b) autumn, notyphoon; (c) summer, typhoon-induced; and (d) summer, no-typhoon. Cross sections were made along the 113.2 E longitude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

### 326 **3.3 O<sub>3</sub> production conditions: comparison of clouds**

327 Clouds efficiently reflect solar radiation (Liou, 1976), and therefore, they have a notable impact on the local formation of  $O_3$ . 328 Figure 6 displays the cross sections of mean ERA-Interim cloud liquid water contents (CLWC) at 14:00 LT of all O<sub>3</sub> pollution 329 days corresponding to the typhoon-induced and no-typhoon scenarios of two seasons, which were also made along the 113.2 °E 330 longitude line, from 26.0 N to 20.0 N (Fig. S4). The comparison of CLWC in the cross sections suggests that typhoons 331 generally resulted in fewer clouds in autumn but more clouds in summer, which agrees well with the comparison of cloud 332 covers in Table 2. The presence of fewer clouds on the typhoon-induced days in autumn can be attributed to two reasons: the 333 influence of dry air masses (indicated by lower RH in Table 2 and Fig. S6) and/or the hindrance of cloud formation by 334 downdrafts. In summer, the strengthened updrafts above the land caused by typhoons favoured cloud formation, which is 335 demonstrated by higher CLWC at the heights of 500-850 hPa and increases in medium and high cloud covers. In areas above 336 the PRD below 850 hPa, downdrafts led to slight decrease of clouds in the typhoon-induced scenario in summer, which is also 337 indicated by reduced low cloud cover. As a consequence of varied cloud covers in each scenario, on average, net surface solar



339 340 341 Figure 5. O<sub>3</sub> 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.

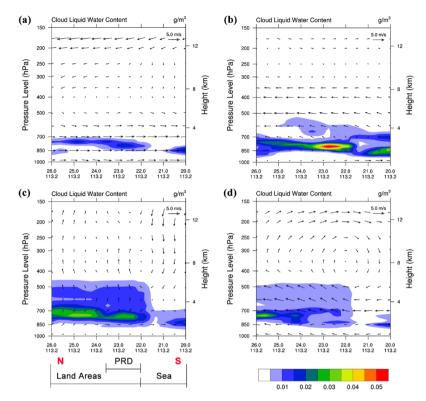




Figure 6. The cross sections of mean cloud liquid water content (g/m<sup>3</sup>) and wind vectors at 14:00 LT for each scenario: (a) autumn, typhoon induced; (b) autumn, no-typhoon; (c) summer, typhoon-induced; and (d) summer, no-typhoon. Cross sections were made along the 113.2 E
 longitude line, from 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.

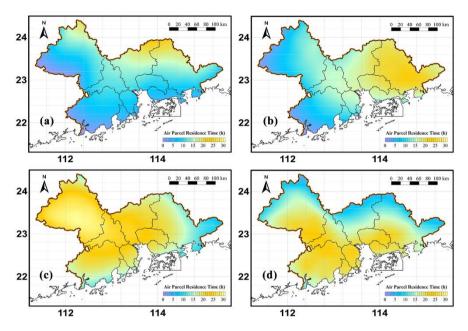
radiation increased by 13% and decreased by 7% on the typhoon-induced days in autumn and summer, respectively (Table 2),

348 which promoted and hindered  $O_3$  production in the PRD during these two seasons, respectively.

# 349 3.4 O<sub>3</sub> accumulation conditions: comparison of APRTs

350 The longer APRTs are, the more likely that  $O_3$  produced by local emissions accumulates within the targeted region and notably 351 contributes to near-ground O<sub>3</sub> pollution. In order to study the effect of typhoons on O<sub>3</sub> accumulation, we calculated APRTs in 352 the PRD in the typhoon-induced and no-typhoon scenarios (Fig. 7) for the further comparisons. On the typhoon-induced days 353 in autumn, APRTs were typically 5-10 hours (mean = 9.5 hours) — shorter than those on the no-typhoon days in autumn 354 (mean = 13.1 hours). In addition, lower APRT values occurred in the central part of the PRD, where high anthropogenic 355 emissions of pollutants are distributed (Zheng et al., 2009). Despite more active O<sub>3</sub> chemistry discussed in the last section, 356 locally sourced O<sub>3</sub> was less likely to accumulate within the PRD in this scenario, potentially limiting the contribution of local 357 emissions for O<sub>3</sub>. The comparison suggests opposite results in the summer scenarios, that is, APRTs on the typhoon-induced 358 days (20–30 hours, mean = 21.0 hours) were overall higher than those on the no-typhoon days (15–25 hours, mean = 16.5359 hours). This favoured the accumulation of locally sourced  $O_3$ , and, to some extent, offset the influence of weakened  $O_3$  formation to maintain high contributions of local emissions to  $O_3$  pollution. Based on the comparison of  $O_3$  production conditions in the previous section and the comparison of  $O_3$  accumulation conditions in this section, typhoons did not provide more favourable conditions for  $O_3$  production and accumulation simultaneously in the PRD in both autumn and summer, thus potentially resulting in a less important role of local contributions in  $O_3$  pollution here. More quantitative evaluations of the contributions from multiple  $O_3$  sources are discussed in Sect. 4.

365



# 366

**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.

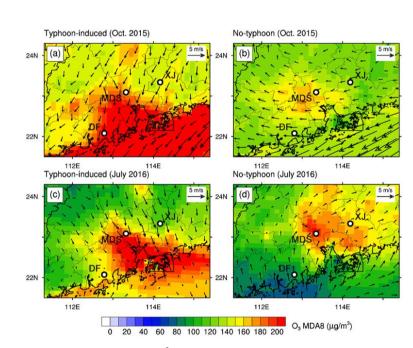
#### 369 **3.5** Meteorological conditions on the close typhoon-induced days

370 On the close typhoon-induced days in the two seasons, stronger north winds prevailed and total cloud cover was higher than 371 that on the no-typhoon days (Table 2), suggesting better conditions for the transport of  $O_3$  but less favourable conditions for 372 O<sub>3</sub> production. As displayed in Fig. S7, the APRT values were significantly lower on the close typhoon-induced days (mean 373 = 6.6 hours, 12.9 hours in autumn and summer, respectively) than on the no-typhoon days, making it even harder for locally 374 sourced  $O_3$  to accumulate within the PRD. Therefore, close typhoons are concluded to promote the transport of  $O_3$  from the 375 outside and to reduce the contributions of O<sub>3</sub> produced from local emissions in a more notable way. In addition, close typhoons 376 led to stronger downdrafts in autumn and updrafts in summer than other scenarios in the same season (Fig. S8). It should be 377 noted that the structure of the summer monsoon cell near the PRD was destroyed in the close typhoon-induced scenario in 378 summer, indicating the stronger influence of typhoons on regional wind fields. The dominant role of  $O_3$  transport during  $O_3$ 379 pollution days in this special scenario agrees well with the reported episode-based analyses (Lam et al., 2005; Li, 2013).

# 380 4 Comparisons of O<sub>3</sub> processes and sources

381 The comparisons of meteorological conditions served as qualitative evidence to determine the general influence of typhoons on O<sub>3</sub> transport, production and accumulation in autumn and summer. Based on the comparison between the CMAO modelling 382 383 results on typical O<sub>3</sub> pollution days in October 2015 and July 2016, more quantitative evidence can be presented. Figure 8 384 displays modelled mean O<sub>3</sub> MDA8 concentrations and wind fields (at 14:00 LT) on the typhoon-induced and no-typhoon O<sub>3</sub> 385 pollution days of two seasons. Large standard-exceedance (>  $160 \mu g/m^3$ ) areas were distributed in the PRD on most days, and 386 the typhoon-induced days of both seasons generally featured higher  $O_3$  levels. The distinct wind fields for these scenarios, 387 which were consistent with those in the longer timespan (Fig. S5), indeed led to different spatial distributions of O<sub>3</sub>. Generally, 388 the most severe O<sub>3</sub> pollution occurred in the downwind areas, such as the central and southern parts of the PRD on the typhoon-389 induced days in October 2015, the central PRD on the no-typhoon days in October 2015, and the northern and eastern PRD on 390 the no-typhoon days in July 2016. On the typhoon-induced days in July 2016, high levels of  $O_3$  accumulated around the PRE. 391 In this section, we discuss the different contributions of various O<sub>3</sub> processes and sources on these days to better understand 392 the effect of typhoons on O<sub>3</sub> pollution in the PRD.

393



394

**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) 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)

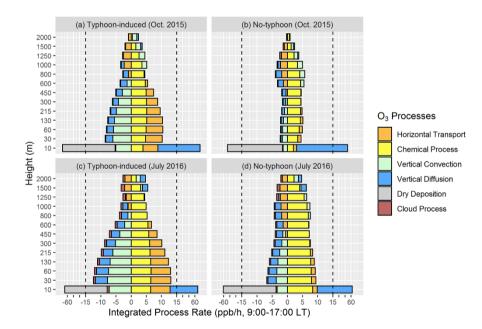
397 November 2015); (c) the typhoon-induced days in July 2016 (7–8 and 30–31 July 2016); and (d) the no-typhoon days in July 2016 (22–26

398 and 29 July 2016). Three representative sites in the PRD are shown as black circles in the plots: XJ, Xijiao; MDS, Modiesha; DF, 399 Duanfen.

# 400 4.1 O<sub>3</sub> processes: transport vs chemical process

401 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) O<sub>3</sub> PA results within the 402 403 PRD in all scenarios share similar characteristics. Dry deposition dominated O<sub>3</sub> removal near the surface, and it also led to 404 high gradients of  $O_3$  concentrations that promote downward  $O_3$  diffusion. Within the PBL (about 0–1 km in height),  $O_3$  was 405 mainly contributed by horizontal transport and chemical process, and vertical convection led to the drop of  $O_3$  concentrations. However, differences existed between the O<sub>3</sub> PA results in the typhoon-induced and no-typhoon scenarios, indicating the 406 407 impact of typhoons on the transport and production of  $O_3$ . In both months, typhoons led to notably higher contribution of 408 horizontal transport to O<sub>3</sub>, especially in the lower and middle part of the PBL. Within the PBL, on average, it increased from -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 409 410 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 411 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 412 413 hindered  $O_3$  production in autumn and summer, respectively. These results agree well with the comparisons of  $O_3$  transport 414 and production conditions in the previous section.

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416

417 Figure 9. The daytime-mean (9:00–17:00 LT) hourly contributions of O<sub>3</sub> processes within the PRD in vertical layers 1–13 on representative

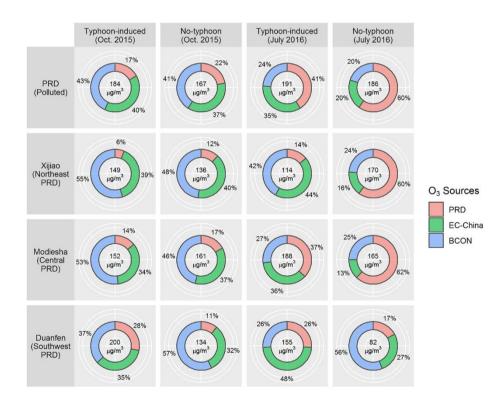
418 O<sub>3</sub> pollution days: (a) the typhoon-induced days in October 2015 (14–16 and 21 October 2015); (b) the no-typhoon days in October 2015

419 (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

420 in July 2016 (22–26 and 29 July 2016).

# 421 4.2 O<sub>3</sub> sources: local sources vs regional sources

422 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) O<sub>3</sub> SA near the ground (about 0–80 m in height) on typhoon-423 424 induced and no-typhoon  $O_3$  pollution days are illustrated in Fig. 10. For polluted regions within the PRD, stronger  $O_3$ 425 production under typhoons did not lead to a higher proportion of local contributions to  $O_3$  pollution in October 2015 — it even 426 decreased from 22% (on the no-typhoon days) to 17% (on the typhoon-induced days). The contributions of EC-China emissions and BCON, in contrast, increased slightly from 37%, 41% to 40%, 43%, respectively. The distinction of the O<sub>3</sub> SA 427 428 results is more apparent for the summer scenarios, that is, typhoons resulted in growing contributions from O<sub>3</sub> transported from 429 other regions (from 40% to 59%) but decreased local contributions (from 60% to 41%) in July 2016. More favourable  $O_3$ 430 accumulation conditions (indicated by higher APRTs on the representative typhoon-induced O<sub>3</sub> pollution days in summer (Fig. 431 S9) were far from sufficient to compensate for the effect of weakened  $O_3$  production on the high contributions of local sources. 432



433

- **Figure 10.** The mean O<sub>3</sub> SA near the ground (about 0–80 m in height) on the represented typhoon-induced and no-typhoon O<sub>3</sub> pollution
- 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
- 437 conditions of the d02 modelling.

438 Furthermore, owing to the variations of wind fields, the comparison results of O<sub>3</sub> SA in different parts of the PRD may differ

439 from the regional ones. For instance, while the comparisons of  $O_3$  SA in the Xijiao and Modiesha site (located in the northeast

440 and central part of the PRD, respectively) agree well with those in the polluted regions of the PRD, higher contributions of 441 PRD emissions for  $O_3$  can be found in the Duanfen site (located in the southwest part of the PRD) on the typhoon-induced 442 days of two months in comparison to these on the corresponding no-typhoon days (Fig. 10). Since the site was located in the 443 downwind region in the typhoon-induced scenario in October 2015 (Fig. 8a), enhanced O<sub>3</sub> production led by typhoons from 444 the massive emissions of  $O_3$  precursors in the central PRD (Zheng et al., 2009) contributed to higher local contributions for  $O_3$ 445 pollution here (as the distribution of local contributions in percentage to daytime  $O_3$  shown in Fig. S10, the highest local contribution in the PRD occurred in areas near the Duanfen site and almost reached 40% in this scenario, which was even 446 447 higher than that in the corresponding no-typhoon scenario (33%)). In the no-typhoon scenario in July 2016, the site was located 448 in the upwind regions under the prevailing of southwest winds, limiting the contributions of local emissions for  $O_3$  at the site 449 (Fig. 8d). Thus, higher local contributions can also be found in the typhoon-induced scenario in this month.

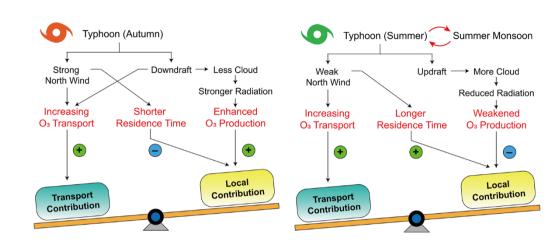
# 450 **5 Discussion and conclusions**

451 The significance of typhoons on  $O_3$  pollution in the PRD calls for thorough evaluations of the different causes of  $O_3$  pollution 452 with the appearance of typhoons in the Northwest Pacific. In this study, we revealed the different impacts of typhoons on  $O_3$ 453 transport, production and accumulation in the PRD (as summarised in Fig. 11) through systematic comparisons of 454 meteorological conditions, the contributions of various  $O_3$  processes and sources in the typhoon-induced and no-typhoon 455 scenarios. We found that typhoons tended to promote  $O_3$  transport towards the PRD, but failed to provide more favourable  $O_3$ 456 production and accumulation conditions simultaneously, which limited the contribution of local emissions to  $O_3$  pollution. 457 Furthermore, there were also differences between the influence of typhoons on  $O_3$  pollution in autumn and summer. More 458 favourable transport conditions occurred in the typhoon-induced scenario in autumn, which was characterised by higher wind 459 speeds and the increased influence of downdrafts. In summer, the mixed types of air masses in the typhoon-induced scenario 460 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, 461 and thus more rapid  $O_3$  production in autumn, but shorter APRTs (5–10 hours) suggest that locally sourced  $O_3$  was hard to 462 accumulate within the PRD. As a result, the contributions in percentage of local emissions to  $O_3$  pollution decreased (slightly 463 464 by ~5% for the polluted regions of the PRD in October 2015). In contrast, in summer, intensified updrafts associated with 465 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 466 (which decreased by ~20% for the polluted regions of the PRD in July 2016). However, due to the variations of wind fields 467 under different scenarios, the changes of local and transport contributions for  $O_3$  led by typhoons were different in the 468 469 southwest part of the PRD, that is, higher contribution from emissions within the PRD and reduced transport contribution 470 occurred in the typhoon-induced scenarios in both seasons. As for the close typhoon-induced scenario, O<sub>3</sub> transport was further

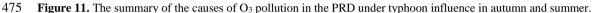
471 strengthened, but meteorological conditions in the PRD became less favourable for both the production and accumulation of

472 O<sub>3</sub>.





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476 The East Asian monsoon, changing with seasons, has a pronounced impact on local meteorological conditions as well as the 477 characteristics of  $O_3$  pollution in East China (He et al., 2008). The seasonal behaviour of the East Asian monsoon is likely to 478 result in the seasonally varied effect of typhoons on  $O_3$  pollution in the PRD. In October, the summer monsoon has almost 479 finished its retraction and the winter monsoon is beginning (Ding, 1994). Thus, there are not many obstacles to the southward 480 movement of typhoon periphery and the transport of  $O_3$  towards the PRD by the continental and coastal air masses. Large 481 downdraft-influenced areas in Central and South China occur in this scenario, and high O<sub>3</sub> levels and low RH in these areas 482 indicate the potentially important role of directly downward  $O_3$  transport. In July, the summer monsoon reaches its strongest 483 (Ding, 1994). The interaction between typhoon periphery and the summer monsoon results in stagnation and enhanced updrafts 484 above the land areas of the PRD and its surroundings. Only when typhoon is close enough to the PRD is the stagnation 485 terminated and the structure of the summer monsoon cell broken. This also explains why some summertime typhoon-induced 486 O<sub>3</sub> episodes in the PRD can be typically divided into two periods, as stagnation leads to the accumulation of locally produced 487 O<sub>3</sub> in the first phase and strong northerly winds strengthen O<sub>3</sub> transport before the landing of typhoons (Lam et al., 2005; Li, 488 2013). It should be noted that updrafts, rather than downdrafts, prevailed on the typhoon-induced  $O_3$  pollution days in summer. 489 High levels of  $O_3$  did not necessarily occur in the regions dominated by downdrafts in this scenario, suggesting a less notable 490 connection between downdrafts and summertime  $O_3$  pollution in the PRD. Further investigations are required to trace the 491 detailed process of downward  $O_3$  transport, including the stratosphere-troposphere exchange (Stohl et al., 2003), in each 492 scenario, and quantify their contributions to near-ground O<sub>3</sub> pollution.

493

494 Some limitations remain in this study. We chose O<sub>3</sub> pollution days as individual samples, ignoring the influence of O<sub>3</sub> pollution 495 on the previous days. Thus, more detailed full-episode analyses are required. Moreover, owing to the small sampling size, the 496 influence of typhoons on  $O_3$  pollution in the PRD is still not fully understood, including, for instance, the detailed connections 497 between the features of typhoons (intensity, position) and O<sub>3</sub> pollution. However, the comparisons of meteorological conditions, 498  $O_3$  processes and sources in different scenarios and seasons demonstrate the complex causes of typhoon-induced  $O_3$  pollution 499 in the PRD — typhoons tend to enhance O<sub>3</sub> transport into the PRD in both seasons, but their impacts on the production and 500 accumulation of  $O_3$  are completely different. As a result, emissions within (outside of) the PRD are likely to contribute less 501 (more) on the typhoon-induced  $O_3$  pollution days than on the no-typhoon days. In order to effectively alleviate  $O_3$  pollution 502 and to reduce the population exposure in the PRD, more attention should be paid to controlling anthropogenic emissions of  $O_3$ 503 precursors on a larger scale, rather than focusing on local emission, under typhoon influence. For air quality management, it is suggested to comprehensively evaluate the efficiency of fractional local and non-local emission reductions to reduce O<sub>3</sub> 504 505 levels in the PRD in different scenarios (Thunis et al., 2019; Thunis et al., 2020). This study also suggests that a thorough 506 evaluation of  $O_3$  transport, production and accumulation conditions can be applied to understand the causes of regional  $O_3$ 507 pollution not only in the PRD, but also in other regions. The results will help find efficient strategies to alleviate regional  $O_3$ 508 pollution as well as to reduce its adverse effects.

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

511

512 Author contributions. KQ, XW and YZ designed the study. KQ, XW, and TX did the simulation work, including the operation 513 of the WRF, SMOKE and CMAQ models. JS, HD, LZ and YZ provided observational results of field campaigns and the 514 routine monitoring datasets for the evaluation of model performance. KQ, XW, YY and YZ analysed the modelling results. 515 KQ, XW, YY and YZ wrote and revised this paper, with critical feedbacks from all other authors.

516

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

518

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