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Spatiotemporal Source Apportionment of Ozone Pollution over the Greater Bay Area

Yiang Chen [1], Xingcheng Lu [2*] Jimmy C.H. Fung [1,3]

1 Division of Environment and Sustainability, The Hong Kong University of Science and Technology, Clear Water Bay, Kowloon, Hong Kong SAR, China

2 Department of Geography and Resource Management, The Chinese University of Hong Kong, Sha Tin, New Territory, Hong Kong SAR, China

3 Department of Mathematics, The Hong Kong University of Science and Technology, Clear Water Bay, Kowloon, Hong Kong SAR, China

Correspondence to: Xingcheng Lu (xingchenglu2011@gmail.com)

Abstract. It has been found that ozone (O_3) pollution episodic case is prone to appear when the Greater Bay Area (GBA) is under the control of typhoons and sub-tropical high-pressure systems in summer. To prevent these pollutions effectively and efficiently, it's essential to understand the contribution of O_3 precursors emitted from different periods and areas under these unfavorable weather conditions. In this study, we further extended the Ozone Source Apportionment Technology (OSAT) from the Comprehensive Air Quality Model with Extensions (CAMx) model to include the function to track the emission periods of O_3 precursors. Then the updated OSAT module was applied to investigate the spatial-temporal contribution of precursors emissions to the O_3 concentration over the GBA in July and August 2016, when several O_3 episodic cases appeared in this period. Overall, the emissions within GBA, from other regions of Guangdong province (GDo), and the neighbouring provinces are the three major contributors, which account for 23%, 15%, and 17% of monthly average O_3 concentration, respectively. More than 70% of O_3 in the current day is mainly formed from the pollutants emitted within 3 days and the same day's emission contributed approximately 30%. During the O_3 episodes, when typhoon approached, more pollutants emitted 2-3 days ago from the GDo and adjacent provinces were transported to the GBA, leading to the increase of O_3 in this region. Under the persistent influence of northerly wind, the pollutants originating from eastern China earlier than 2 days ago can also show an obvious impact on the O_3 over the GBA in the present day, accounting for approximately 12%. On the other hand, the O_3 pollution is primarily attributed to the local emission within 2 days when the GBA is mainly under the influence of the sub-tropical high-pressure systems. These results indicated the necessity to consider the influence of meteorological conditions in implementing the control measures. Meanwhile, analogous relationships between source area/time and receptor were derived by the zero-out method, supporting the validity of the updated OSAT module. Our approach and findings could offer more spatial-temporal information about the sources of O_3 pollutions, which could aid in the development of effective and timely control policies.

1. Introduction

As one of the major air pollutants, ozone (O_3) is a secondary pollutant formed by the photochemical reactions of nitrogen oxides (NO_x) and volatile organic compounds (VOCs) in the presence of solar radiation. The surface O_3 has detrimental effects on human health, such as causing respiratory and cardiovascular problems (Maji et al., 2019; Yin et al., 2017). It could also lead to the reduction of crop yield and the damage of vegetation (Gong et al., 2021; Wang et al., 2022c). With the implementing of a series of control policies in China since 2013, the concentrations of other air pollutants, including particulate matter with aerodynamic diameters less than 2.5μm ($PM_{2.5}$), NO_x, and sulfur dioxide (SO_2), have gradually decreased. In contrast, due to the large reduction of NO_x emission and less control of VOCs emission in the early stage of the control period (Liu et al., 2023), the O_3 concentration still continuously increased and has become the major air pollutant across China. The Greater Bay Area (GBA), including nine cities in the Pearl River Delta (PRD) region, Hong Kong (HK), and Macau Special Administrative Regions (SAR), is one of the most developed agglomerations in China and also facing with the heavy O_3 pollution problem. Based on the analysis of surface monitor observation, Cao et al. (2024) and Feng et al. (2023) found that the daily maximum 8hr average (MDA8 O_3) in the PRD region and HK showed an overall upward trend by 1.11 and 0.22 ppbv/year from 2013 to 2019 and from 2011 to 2022, respectively.

The formation of O_3 is closely related to the source of its precursors and much effort has been devoted to investigating the source region and source category of O_3 in the GBA using different methods (Liu et al., 2020a). He et al. (2019) applied the positive matrix factorization (PMF) method to resolve the anthropogenic sources of VOCs. Combining with a photochemical box model with the master chemical mechanism (PBM-MCM), they found that vehicular was the most significant source to the O_3 formation, followed by biomass burning and solvent usage. Li et al. (2012) applied the CAMx-OSAT numerical model to track the source contribution to O_3 in the GBA region and found that elevated local and regional contribution is dominant under the O_3 episodes. Yang et al. (2019b) applied the NAQPMS model with an on-line source apportionment module to explore the source of O_3 in different seasons in the PRD region. Their results shows that the mobile is the largest contributor, followed by industry. Fang et al. (2021) used multi-modelling source apportionments to quantify the source impact on O_3 in the PRD region. The on-road mobile and industrial process were found as two major contribution sections. Integrating satellite data and sensitivity model simulations, Wang et al. (2022a) found that enhanced biogenic emission and cross-regional transport by approaching typhoons are significant factors lead to the ozone pollution in the PRD and Yangtze Rivel Delta (YRD) regions. In addition to the source region and category, emitting time of pollutants is also an important perspective that needs to be better understood for effective and efficient control policymaking. Some studies have attempted to evaluate this temporal perspective (Xie et al., 2021; Ying et al., 2021). Xie et al. (2023) analysed the age evolution of $PM_{2.5}$ during a haze event in eastern China. It showed that during the regional transport stage, more aged particles from the North China Plain (NCP) were transported to the downwind YRD region and leading to a sharp increase in the average age of different components of $PM_{2.5}$ in YRD. Chen et al. (2022c) analyses the temporal contributions of emissions to the concentration of $PM_{2.5}$ in the PRD region and found that pollutants emitted from 2 days earlier were trapped within the PRD region due to the weak wind during the episodic pollution. However, these studies mainly focus on the $PM_{2.5}$ and the temporal contribution of sources to the O_3 in the GBA region still remain unclear.

Besides emission, the meteorological condition, another key factor that can affect the transportation, production, and destruction of O_3 and its precursors, also received much attention and has been extensively studied (Lu et al., 2019; Wang et al., 2017; 2022b). The long/short-term effects of changes in meteorological conditions on ozone concentrations have been investigated through a variety of methods, such as statistical analysis of observations and numerical modelling (Yang et al., 2019a; Xu et al., 2023a; Zheng et al., 2023). Liu and Wang (2020b) conducted sensitivity simulations by the CMAQ model to evaluate the contribution of weather condition variation to summer O_3 levels from 2013-2017. Their results show that the meteorological conditions were more conducive to ozone formation from 2014 to 2016 than in 2013, and it can lead to an increase of more than 10 ppbv in MDA8 O_3 in Guangzhou. Different objective and subjective classification technologies have been applied to summarize the impacts of unfavorable weather patterns on O_3 pollution (Han et al., 2020; Chen et al., 2022b; Cao et al., 2023). Gao et al. (2018) summarized the commonly synoptic patterns in the Guangdong province that O_3 pollution always occurred and concluded that the sub-tropical high-pressure system and typhoons are two major patterns accounting for more than 60% in the PRD regions during 2014 – 2016. The major influence factor and the dominant contributed physical and chemical process were also identified and analyzed (Gong et al., 2022; Zeren et al., 2022; Wu et al., 2023). Ouyang et al. (2022) analysed the impact of a subtropical high and a typhoon on ozone pollution in the PRD region and found that low relative humidity, high boundary layer height, weak northerly surface wind, and strong downdrafts were the main meteorological factors contributing to the pollution. Deng et al. (2019) illustrated that the actinic flux is the important cause of the co-occurrence of high ozone and aerosol pollution under the control of typhoon periphery. Li et al. (2022) also investigated the impact of peripheral circulation characteristics of typhoons and found that the chemical formation and vertical mixing effects are two major contributors to the enhancement of O_3 levels, while the advection showed negative values. Qu et al. (2021) analysed the typhoon-induced and non-typhoon O_3 events in the PRD region and revealed that under the influence of typhoons, the contributions from the transport processes and sources outside the PRD increased. Usually, the ozone events are attributed to changes in meteorological conditions rather than sudden increases in emission intensity (Lin et al., 2019; Xu et al., 2023b). The change of weather conditions will affect the time-sensitivity of emitted pollutants and lead to different types of O_3 pollution that can result from long-range transport of elder pollutants or accumulation of local fresh pollutants. Hence, it's of great importance to clarify the impact of the pollutants from different source areas and emitting periods on the O_3 pollution under different weather conditions in the GBA.

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In this study, the CAMx-OSAT model was extended and used to track the temporal contribution of pollutants to the O_3 pollutions over the GBA under the impact of typhoons and sub-tropical high pressure during July and August in 2016, the two most important weather system that influence O_3 pollutions over the GBA. The rest of this paper is organized as follow. The temporal source apportionment (TSA) method, the configuration of experiments, and the ozone episodes were introduced in section 2. The spatial-temporal source apportionment results and zero-out simulation results were shown and discussed in section 3. The major conclusions were summarised in section 4.

2. Methodology and Data

2.1 Temporal Source Apportionment Method

Previously, we have successfully implemented the $PM_{2.5}$ temporal source apportionment (TSA) method in the Comprehensive Air Quality Model with Extensions (CAMx) model and applied it to investigate the temporal influence of emissions on $PM_{2.5}$ in the GBA (Chen et al., 2022c). Here, we further extend this method to track the temporal contribution of emissions to O_3 and its precursors. The basic mechanism of the TSA method is to track the contribution of pollutants from different emitting periods using a set of tracers. In the TSA method (Fig. 1), the *Precursor Tracer Day-x* was used to track the precursors emitted from *x* days ago. The *O_3 Tracer Day-x* was used to track the O_3 formed from the precursors emitted from corresponding *x* days ago (namely *Precursor Tracer Day-x*). The tracers in *Day-x* can be set into different finer periods (e.g., every 1 hour, 6 hours, 24 hours) as required. The total tracer number will be decided according to the whole tracking period and the minimum tracking period per tracer. For instance, if the whole tracking period is 5 days and the minimum tracking period per tracer is every 6 hours, the total tracer number will be 20. As shown in the Figure 1, during each day's simulation, the contribution of present day's emission will always be tracked by the Day-0 tracers. After completing the current day's simulation and before starting the next day's simulation, each tracer *Day-x*'s value transfers to the corresponding tracer *Day-(x+1)*, which represents one day earlier than *Day-x*, in the following sequence. For example, beginning from the penultimate tracer, namely values in Day-3 transfer and add into Day-4, then the values in Day-2 transfer to Day-3, followed by Day-1 to Day-2, and lastly Day-0 to Day-1 (Dash arrow in Figure 1). Here, the value in Day-3 tracer will add into the last tracer (Day-4) because the last tracer represents the total contribution of pollutants emitted earlier than 3 days ago. More details of this method can be found in Chen et al. (2022c).

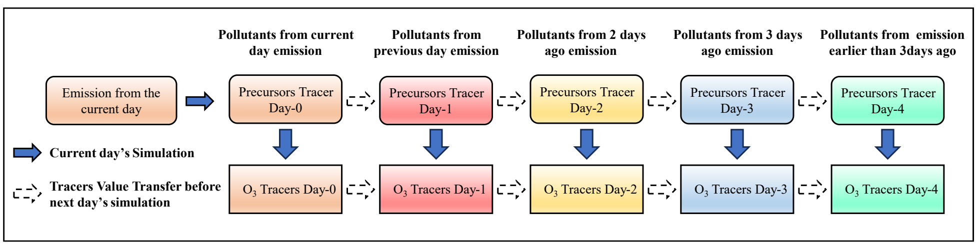

Figure 1. Schematic diagram of temporal source apportionment (colors represent the pollutants released or formed by emissions on different days).

2.2 Model Configuration and Evaluation

The Weather Research and Forecasting (WRFv3.9) model was applied for meteorological field simulation. The initial and boundary condition for WRF model was gained from the Final Operational Global Analysis data (FNL). The CAMx v7.1 was used to simulate the spatial-temporal variation of air pollutants. The initial and boundary condition for CAMx model was provided by the Model for Ozone and Related chemical Tracers, version 4 (MOZART-4). Regarding the emission, a highly resolved emission inventory provided by Hong Kong Environmental Protection Department was used for the GBA region, and the Multi-resolution Emission Inventory for China (MEIC, Li et al., 2017) developed by the Tsinghua University was applied for the area outside the GBA region. The biogenic emission was calculated the Model of Emissions of Gases and Aerosols from Nature (MEGAN version 3.1). The CB05 gas phase chemistry, the ISORROPIA inorganic aerosol scheme and the SOAP secondary organic aerosol scheme were used in the simulation. This model system has been applied to analyse the

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source of O_3, NO_x and $PM_{2.5}$ in the GBA region in the previous studies (Lu et al., 2016; Chen et al., 2022a; Chen et al., 2022c). More configuration of this model system can refer to the work of Lu et al. (2016).

The three-nested simulation domain of the WRF-CAMx model was shown in Figure S1. The resolution of three domains was 27km, 9km, and 3km, respectively. For the source apportionment experiments, the simulation domain was divided into 12 source regions as shown in Figure 2, including North China Plain (*NCP*), eastern China(*EC*), southern western China (*SWC*), other regions of inland China (*Other 2*), ocean and other countries (*Other 1*), neighbouring provinces around Guangdong province (*Neighbor*), Other region within Guangdong province(*GDo*), Guangzhou and Foshan(*GF*), Shenzhen and Dongguan(*SD*), Hong Kong (*HK*), Zhuhai, Zhongshan and Jiangmen (*ZZJ*), Zhaoqing and Huizhou(*GBAo*). The cities within the GBA were separated into different sub-regions mainly based on their geographical location, same as the work of Chen et al. (2022c). Since the source contribution to the O_3 in Zhaoqing and Huizhou is relatively different with that of their neighboring cities (Chen et al., 2022a), these two cities were grouped into one sub-region. The contribution of initial and D1 boundary conditions were also treated as two sources. In the following analysis, for the O_3 concentrations in the target area over the GBA, the influence of pollutants emitted within the target area is treated as the local contribution, and the influence of pollutants originating from the other areas within the GBA region is treated as the regional contribution. The source tracking time period is 5 days (Day-0, Day-1, Day-2, Day3 represent the pollutants emitted within the present day, the previous day, two days ago, and three days ago, respectively. Day-4 the total contribution of pollutants emitted earlier than three days ago). The simulation period is July and August 2016, and the model was spin-up for 7 days to reduce the influence of initial condition.

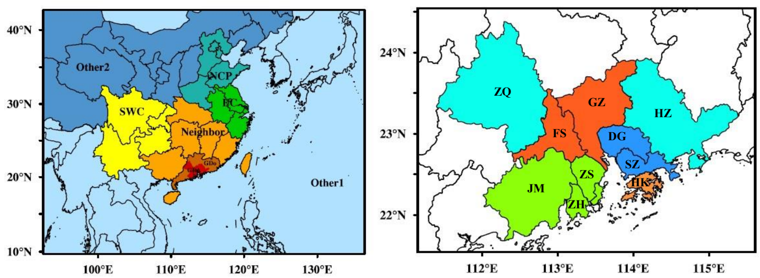

Figure 2. The configuration of source areas in the source apportionment experiments (One color represents one source area. The GBA source were divided into five source areas. *Other 1* represents ocean and other countries. *Other 2* represents other area within the mainland China in the simulation domain.)

The performance of simulated 2-m temperature, 10-m wind speed, and O_3 concentration were evaluated and shown in Table S1. The recommended values suggested by Emery et al. (2001) and EPA (2007) were used as benchmark and shown in the brackets in Table S1. The temperature was a little overestimated with a mean bias (MB) of 0.33, while the wind speed was underestimated with MB of -0.45. The index of agreement (IOA) was 0.82 and 0.70 for temperature and wind speed, respectively. The MBs and IOAs both fulfilled the criteria. But the root mean square error (RMSE) show a little higher than the value of criteria. Regarding the O_3, the IOA reach 0.81. The small positive MB indicated that the model slightly overestimated the O_3 concentration. The normalized mean bias (NMB) is 0.13, which also meet the criteria. The time series comparison (Fig. S2) of average O_3 concentration in Guangzhou, Hong Kong and Zhuhai illustrates that the model can well catch and reproduce the variation trend of O_3 concentration in GBA, although there is a few difference between the simulated and measured concentration for some peaks, like the period between 25 July and 31 July in Guangzhou. Overall, the performance of model simulation is comparable to the other studies in this region (Li et al. 2022; Yang and Zhao, 2023). Therefore, the simulation result is reasonable and can be further used for source analysis.

2.3 Ozone Episodes

There were several O_3 episodes occurred during the simulation period. Here, the 8-h maximum O_3 concentration (MDA8) over the GBA was calculated using the observation data from the surface monitors stations (Fig. 3). The first O_3 pollution occurred between the 7^{th} and 10^{th} of July (Ep1). During this period, the GBA region was firstly controlled by the sub-tropical high-pressure system. When the typhoon north-westerly moved from the east sea area of the Philippines to Taiwan province, the GBA was located in the peripheral subsidence region. After the

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typhoon made landfall, the high-pressure situation in the GBA was relieved and the O_3 concentration decreased. There were another two O_3 episodes between 24 July and 1st August. The GBA was mainly influenced by the sub-tropical high-pressure system during 24th-26th July (Ep 2), while the synoptic condition of GBA between 30th July-1st August (Ep3) was similar to that of Ep1. During the Ep3 period, there was another typhoon moving north-westerly from the east sea area of the Philippines and influencing the GBA region. It was found that this type of typhoon movement path was often accompanied by the occurrences of O_3 pollution in the GBA (Wang et al., 2022a). In late August, under the joint influence of the subtropical high-pressure system and the typhoon, the O_3 over the GBA maintained a high concentration level between the 21st -31st of August (Ep4). Unlike the moving paths of the previous two typhoons, this typhoon was moving southerly from the sea areas south of Japan and stayed near the sea areas east of Taiwan province. The typhoon moved north after 27th August, and northerly winds prevailed in the GBA. Hence, we conducted the simulation of O_3 concentration in the GBA during July-August 2016 and analysed the spatiotemporal contributions of emissions in these episodic cases.

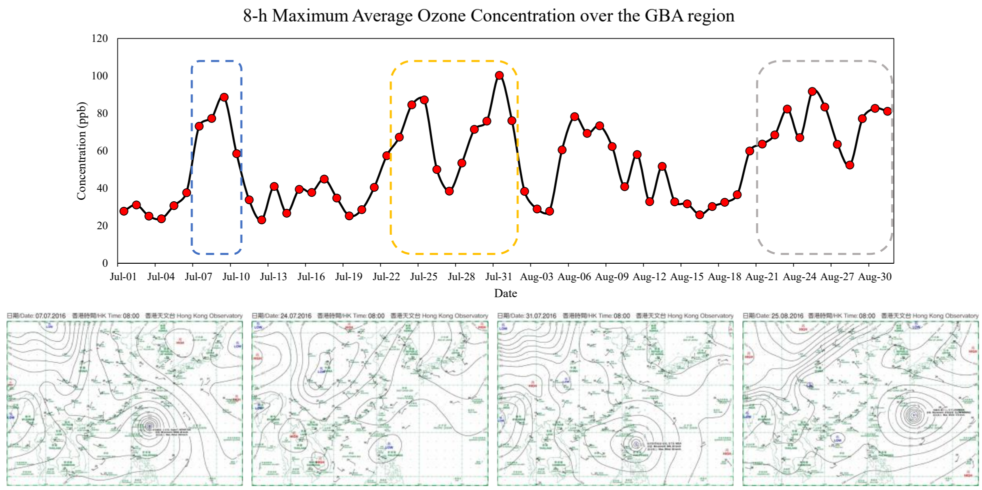

Figure 3. The time-series of the MDA8 O_3 concentration over the GBA during July-August 2016 and the synoptic pattens during the O_3 episodes. (The weather charts were downloaded from the Hong Kong Observatory; https://www.hko.gov.hk/en/wxinfo/currwx/wxcht.htm)

3. Result and Discussion

3.1 Source Area Contributions

The contribution of different source areas to the average O_3 in the GBA region was shown in Table 1. Here, the contribution from initial and boundary condition were treated as background contribution. Regarding the monthly average O_3 concentration over the GBA region, the emission within the GBA can contributed about 23%. The pollutants from other regions within Guangdong Province (GDo) and neighbouring provinces also have large contribution, accounting for approximately 15% and 17%, respectively. Under the influence of prevailing south winds in the summertime, the contribution from ocean and other countries can also account for about 20%. As some studies suggested that O_3 originating from foreign countries is quite limited (Sahu et al., 2021), the main contributor of this source is likely to be marine ship emissions from ocean. The pollutants from other source regions have limited effect on the O_3 in the GBA.

The monthly average source area contribution to four sub-regions within the GBA region can be found in Table S2. Results shows that the local emission has large influence on O_3 in GF and SD regions, accounting for 17% of O_3 but it has impact lower than 10% on O_3 in ZZJ region and HK city. The contribution of GBA regional emission (contributed by other GBA tagged regions) has a relatively larger impact to the monthly average O_3 concentration in GF region than the other sub-regions. It's because the prevailing southerly wind in summer, so the pollutants within the GBA region have a large influence on O_3 in GF area. The influences of pollutants from GDo and

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neighboring to different subregions ranges from 25% to 31%. As the coastal regions, the ZZJ region and HK city also suffer more from sources of ocean and other countries, which occupied about 24% and 27%.

Regarding the average O_3 concentration over the GBA region in different episode periods, it can be found that, during the typhoon episodes (i.e., Ep1, Ep3 and Ep4), the contribution of non-local emission has increased. The typhoon paths are quite similar in the Ep1 and Ep3 episodes (Fig. S3). Results show that the total contribution of GDo and neighbouring provinces have increased and reached more than 50% for O_3 over the GBA in these two typhoon episodes. As shown in Figure S4, with the approaching of typhoon, the wind speed increased and the average wind direction over the GBA changed from south to north. Therefore, more pollutants from the surrounding provinces were transported to GBA. Considering the typical circulation of typhoon periphery (Figure. S4 and S6), it was judged that more pollutants may come from Jiangxi, Fujian, and Hunan provinces. During the Ep1 and Ep3 episodes, the contribution of local emission in different sub-regions slightly decreased. With the change of the wind direction from south to north in these two periods, the influence of pollutants within GBA to O_3 in GF area decreased from 15% to 8%. The contribution of GBA emission to the O_3 in other sub-regions increased, especially the ZZJ area and HK city. It is because with the change of wind direction, these two regions were located at the downwind area of the GF and SD regions, which are the emission hotspots within the GBA. At the same time, the contribution of source from ocean and other countries also decreased approximately 10%. The influence of GDo and neighbouring provinces increased 27%, 21%, 32% and 22% for GF, SD, ZZJ regions and HK city, respectively.

In another typhoon process (Ep4), where the typhoon's moving path differed from the other two typhoon cases, there was an increase in contribution from GDo and neighbouring provinces under the influence of persistent northerly wind. Furthermore, it was observed that pollutants from eastern China (EC) and North China Plain (NCP) could also influence the O_3 levels in the GBA, accounting for approximately 12%. Similar increases in the impact of emissions from the EC and NCP were also found in the four sub-regions.

In the Ep2, the GBA was mainly controlled by the sub-tropical high-pressure system and southerly wind still prevailed. However, the wind speed was low and conducive to the accumulation of the pollutants. Hence, the local sources were the dominant contributor and accounting for about 44% but the contribution from GDo and neighboring provinces decreased. For O_3 in the GF region, as discussed above, the O_3 in the GF regions is more susceptible to emissions within the GBA under the prevailing southerly wind. Thus, not only the local contribution but also the GBA regional contribution largely increased in the GF region. The regional contribution is larger in the GF region, increasing from 15% to 33%. For the other sub-regions, the main increase was in local contributions.

Table 1. Contribution of pollutants from different source areas to the O_3 concentration over the GBA in different cases.

Case	GBA	GDo	Neighbor	Other 1	EC	SWC	NCP	Other 2	Background
Monthly	23%	15%	17%	20%	3%	1%	1%	1%	20%
Ep1	18%	21%	35%	10%	3%	0%	0%	0%	13%
Ep2	44%	11%	7%	27%	0%	0%	0%	0%	11%
Ep3	19%	34%	25%	9%	3%	0%	1%	1%	9%
Ep4	20%	16%	18%	15%	8%	1%	4%	3%	14%

* Here, *GDo* represents areas outside the GBA but within Guangdong province. *Neighbor* represents the provinces around Guangdong province. *Other 1* represents ocean and other countries. *Other 2* represents other areas within the mainland China in the simulation domain. *Background* represents the contribution of initial and boundary conditions.

3.2 Emission Period Contributions

The contribution of pollutant emitted from different time periods to the average O_3 in the GBA and sub-regions was shown Figure. 4 and Table. S3. The background contribution was not considered in the temporal source contribution analysis. This is because the background contribution is primarily derived from boundary conditions, and its temporal contribution was calculated based on the time when the pollutants were transported into D1 rather than the actual emission time.

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Overall, under the general monthly condition, the emission within 3 days (namely from Day-0 to Day-2) account for approximately 73% of monthly average O_3 concentration within GBA. The largest proportion of O_3, around 31%, formed from the current day's emission (Day-0) and the contribution of pollutants from earlier emission periods decreases as time increases. For the monthly average O_3 in different sub-regions, more O_3 in GF and SD regions was formed from the emission from Day-0, which contributed about 37% and 36%, respectively. The contribution of emissions from Day-1 decreased to about 23% in these two regions. The contribution of Day-0 and Day-1 emissions was relatively small but stable for HK city and ZZJ region, which account for around 25% and 27%, respectively. The influence of pollutants emitting earlier than 3 days ago (i.e., Day-4) is mostly lower than 20%.

The situations are different during the pollution periods. The contribution of emission from the current days to the average O_3 over the GBA both decreased in two typhoon cases with similar moving path (Ep1 and Ep3), but the contribution of emissions from Day-1 to Day-3 increased 14% and 8%, respectively. And the influence of pollutants emitted earlier than 3 days ago (>=Day-4) decreased 11% in Ep1 and almost no change in Ep3. This indicates that these two ozone pollutions were caused by the accumulation of pollutants within the current 3 days.

For another typhoon case (Ep4), the contribution from the Day-0 decreased approximately by 11%, comparing with the monthly contribution over the GBA. At the same time, the influence of pollutants from earlier emitting period increased, especially for those earlier than 3 days ago (Day-4). It means that the O_3 pollution for this period is a persistent pollution process. The major contributor should not only the local emission, but also the long-range transport. Similar variation trend of the temporal contributions of emission to different sub-regions can be concluded, which also illustrated that the O_3 pollution is usually a regional problem.

For Ep2, the contribution of emissions from Day-0 increased approximately 18%, while the influence of Day-1-3's emissions decreased about 18%. According to the source area contribution result, the source area of O_3 over GBA in Ep2 is mainly local sources. So the contribution of the freshly emitted pollutants was larger. The contribution of Day-4 emissions to HK and ZZJ regions in Ep2 is larger. It is probably because the prevailing south wind direction brought more airflow came from the ocean. Compared with the emission of GF and SD regions, HK city and ZZJ region have lower emission amounts. At the same time, HK city and ZZJ region locate in the upwind region, and the pollutants from GBA would have a smaller influence on the O_3 in these two regions. Hence the fresh pollutants amount was smaller and contributed similarly to the Day-4 emissions, which is an accumulated amount.

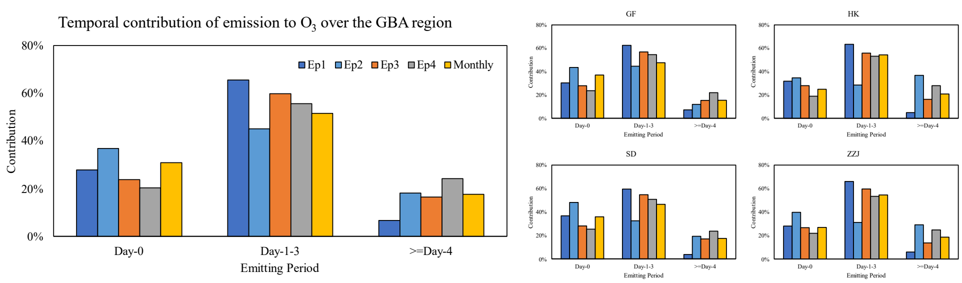

Figure 4. Contribution of pollutants from different emitting periods to the O_3 concentration over the GBA in different cases.

3.3 Source Area-Time Contributions

To further clarify the relationship between sources and the O_3 concentration of target regions, the evolution of O_3 from various source areas and periods were analyzed. Figure 5 shows the time series of the contributions from different source areas and precursors emission periods for the average O_3 concentration in the GBA region.

Regarding the monthly average O_3 concentration over the GBA, the emissions within the GBA is the major contributor and generally have a larger effect in the current day. Under the control of southerly wind, as shown in Figure 6, the pollutants emitted 1 day ago (Day-1) were gradually transported out of the GBA, and the influence

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of the GBA's emission earlier than Day-1 is much lower. At the same time, the pollutants of GDo and neiboring provinces emitted 1 day ago began to have impact on the O_3 in the GBA. However, the elder pollutants from GDo and neighboring provinces cannot greatly influence on the O_3 in the GBA due to the prevailing southerly wind.

However, regarding the O_3 pollution between 7th and 10th July (Ep1), the major contributors changed. On 7th July, the GBA was under the control of the subtropical high system, and the typhoon was located near the east of Taiwan province. The weather condition was unfavourable for pollutants dispersion, and the O_3 sourced from Day-1 emission within Guangdong provinces was trapped. The prevailing wind shifted to northerly wind, and it also brought some elder pollutants from neighboring provinces to the GBA. With the approach of the typhoon on 8th – 10th July, although stronger northwest wind speeded up the diffusion of pollutant from GBA and deceased the local contribution, it also transported more elder pollutants from the northern inland to the GBA. It can be found that the emission from GDo in the present day also had a significant contribution. At the same time, the pollutants from the neighboring provinces dominated the Day-1 to Day-3 emissions. Moreover, the pollutants emitting 2 days ago in the eastern China (EC) region were also transported southerly and affected on the O_3 of GBA in the current day. Figure 7 shows the spatial distribution of average source contribution during the Ep1 period. Compared with the monthly average (Figure 6), it was found that the elder pollutants originating from the GBA can be transported back and influence the O_3 concentration in the western part of GBA during the Ep1 period. This is because easterly and east winds blew over the GBA from 5th-6th July (Before Ep1, Figure S4). The pollutants emitted within the GBA were transported to northwest inland. However, under the influence of northwest wind, they were transported back to the GBA again. It can also be seen that the pollutants from the GDo 1 day ago were transported downwind quickly, contributing to a high O_3 concentration over the Pearl River Estuary. According to the wind pattern, they mainly came from the northern and western parts of the Guangdong province. Meanwhile, the neighboring provinces' emissions from Day-1 to Day-3 were also transported to the GBA with the northwest wind, continuously affecting the O_3 over this region.

For the Ep3 O_3 pollution process, results show that the pollutants from GDo and neighboring provinces were also the major contributors. From 30th– 31st July, the GBA was under the control of high pressure, and it blew weak north wind in this region. Afterward, the approaching of typhoon (1st August) further strengthened the cross-regional transport of pollutants. The difference between Ep3 and Ep1 is that the emissions from GDo have a larger impact on the Day-1 and Day-2 emissions. Additionally, while pollutants from neighboring provinces and EC in Day-4 emission only accounted for about 5ppb in Ep1, they can still contribute to about 10ppb in Ep3. The possible reason is that northerly wind prevailed over Fujian, Jiangxi, and Hunan provinces during the whole Ep1 period (Figure S4), while easterly wind still blew over these provinces during the earlier period of the Ep3 (30th – 31st July, Figure S6), which slowed the transport and influence of pollutants from the neighboring provinces. Generally, the pathways of typhoons in the Ep1 and Ep3 episodes were quite similar, and the influence regions of typhoon wind field mainly cover Guangdong and neighboring provinces. Therefore, the major source area and source time are quite similar in these two cases. To prevent this type of O_3 pollution, earlier emission control (at least 3 days ago) and collaboration with neighboring provinces will gain a better control result.

On the other hand, the situation is different for the Ep2 ozone pollution. Under the control of high-pressure system and weak southerly wind (Figure S5), the major contributors were mainly the pollutants from the GBA and the ocean. Unlike the Ep1 and Ep3, the pollutant emitted within the GBA still dominant in the Day-1's emission contribution. Under the influence of southerly wind, there is no large amount of pollutants migration from north inland to the GBA and the local pollutants were slowly moving out the GBA. Thus, the pollutant emitted earlier than 2 days ago (>=Day-2) have a smaller contribution. As shown in Figure 8, the overall diffusion of pollutants within the Guangdong province is much slower during Ep2. The contribution of GBA emissions can still reach more than 10 ppb in the Day-1 emission. These results indicated that this pollution process was mainly driven by the local pollutants within the current 2 days. Hence, emission control should focus on the local sources, and 1-2 days in advance is more efficient.

For the last O_3 pollution process (Ep4), from the 21st to 25th August, the eastern and southern China were mainly control by the sub-tropical high-pressure system. At the same time, under the joint influence of peripheral subsidence airflow of typhoon, the wind speed over this region was slow (Figure S7). The weak wind not only trapped the O_3 formed local emission but also the O_3 formed from cross-regional transported pollutants. The pollutants from GBA sources mainly dominated the Day-0 and Day-1 emission's contribution, while Day-2 and Day-3 emissions mainly consisted of pollutants from GDo and neighboring provinces. After that, the typhoon moved northerly and the stronger northly wind further broaden the source areas of the O_3 in the GBA (Figure S7).

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The major contributor of Day-2 and earlier periods' emission changed to pollutants from the EC and NCP regions. The pollutants emitted earlier than 2 days ago from EC have an important contribution, which accounted for about 12%. Furthermore, the pollutants emitted 3 days ago from the North China Plain (NCP) can also have an obvious impact on O_3 over the GBA from July 28th- 30th, which can be up to 10%. Hence, to prevent the occurrence of this pollution, the emission control region should be further broadened and continuously implemented as it lasted for a longer period compared with the other three pollutions.

Figure S8 shows the time series of the contributions from different source areas and precursors emission periods for the average O_3 concentration in the GF region and HK city. GF region is located at the inland of the GBA. It is the emission hotpot of the GBA with higher O_3 concentration (Chen et al., 2022a). HK city located at the mouth of the PRD. According to previous source apportionment studies (Li et al., 2012, 2013), the pollution in HK city is more attributed to the emissions outside the GBA compared to the other cities of the GBA. Regarding the O_3 in GF region, Day-0 emission was usually contributed by the local emission and the regional transport within the GBA, which have a similar contribution. The major source areas of the Day-2 to Day-4 emissions contributing to the O_3 in GF in different episodic cases varied similarly to the ones contributing to the average O_3 in the GBA. Generally, the influence of local and GBA regional pollutants to O_3 in the GF region decreased quickly within 1 day. However, the regional emission can still have important contribution in the episodic case that southerly wind blew, such as the 24th -25th July (about 26%) in the Ep2 and 23rd -25th (about 15%) in the Ep4. For the O_3 in HK city, the local emission amount is low, and its impact is also limited to the current day. In addition, the O_3 in HK city is also susceptible to the impact of pollutants from the ocean but less from the GBA regional emissions. During the Ep1 periods, it was observed that the contribution of the GBA regional source largely increased in the Day-0 emission as the prevailing wind direction shifted to north. On the other hand, neighboring provinces' emissions dominate the contribution of emissions from Day-1 to Day-3. Unlike the GF region, the influence of EC emission on the O_3 in HK is also limited in Ep1. Similar conclusions can be drawn for the evolution of the spatiotemporal contribution of emission in Ep3. As discussed above, the O_3 pollution in Ep2 is mainly driven by local emissions. Thus, the O_3 concentration in HK city, which is in the upwind region with small local emissions, is much lower than the O_3 concentration in the GF region. In Ep4, same as the GBA average and GF region, the impact of pollutants from EC and NCP became important in the Day-2 and Day-3 emissions, which can contribute up to 20% of O_3. These results indicate that although O_3 is usually a regional pollution problem, it's necessary to consider the local characteristics of different sub-regions while making more specific prevention and control policies.

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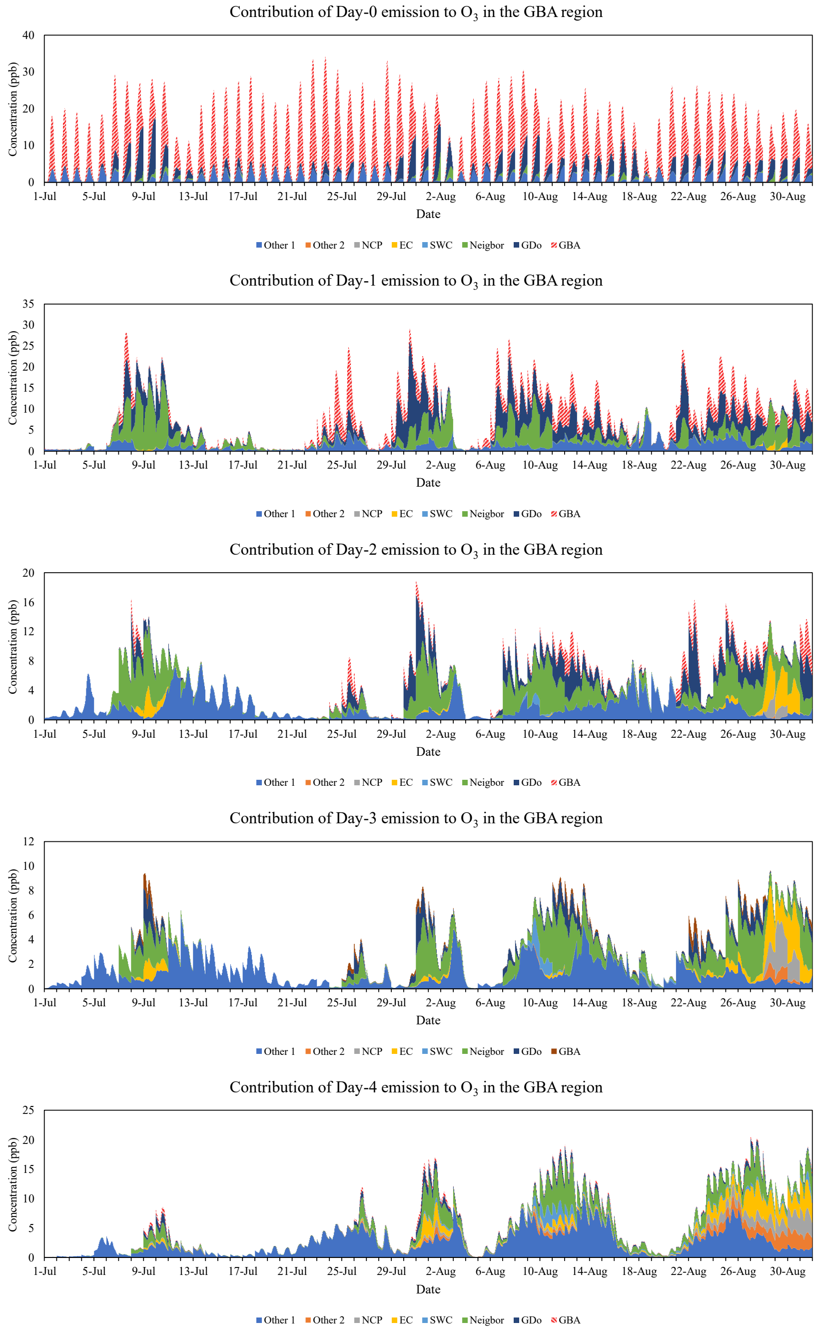

Figure 5. Time series of contributions from different source areas and emitting periods to the O_3 concentrations in the GBA. (*GDo* represents areas outside the GBA region but within Guangdong province. *Neighbor* represents the provinces around Guangdong province. *Other 1* represents ocean and other countries. *Other 2* represents other area within the mainland China in the simulation domain.)

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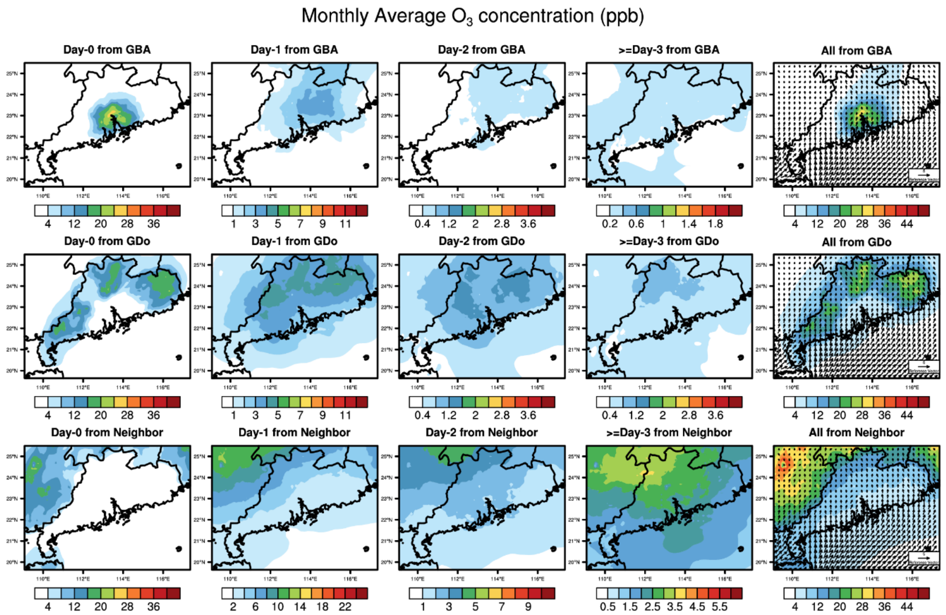

Figure 6. Spatial distribution of monthly average O_3 concentration between 9:00-17:00 (Local time) contributed by emission of GBA, other regions within Guangdong province (GDo), and neighboring provinces (Neighbor) from various periods. (Unit: ppb. Due to the large variation of contribution, the colorbar range of each sub-figure is different)

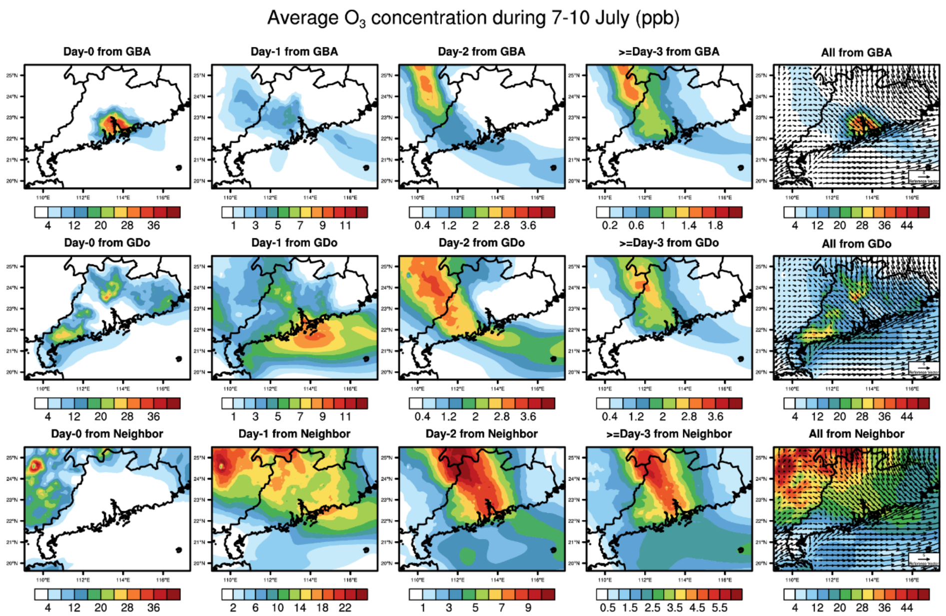

Figure 7. Spatial distribution of average O_3 concentration between 9:00-17:00 (Local time) on 7th-10th July 2016 contributed by emission of GBA, other regions within Guangdong province (GDo), and neighboring provinces (Neighbor) from various periods. (Unit:ppb. The colorbar range of each sub-figure is same as the one in Figure 6)

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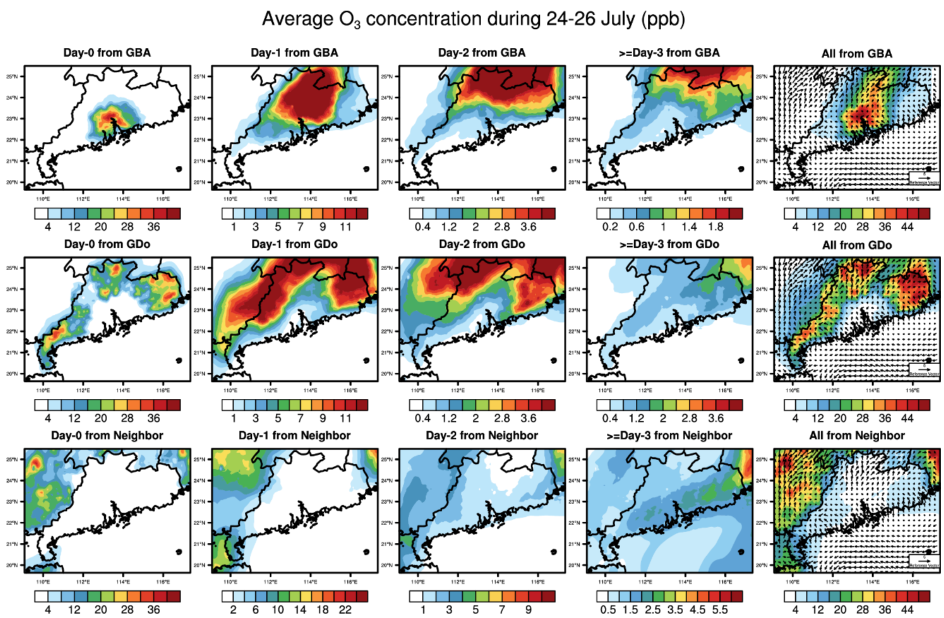

Figure 8. Spatial distribution of average O_3 concentration between 9:00-17:00 (Local time) on 24th-26th July 2016 contributed by emission of GBA, other regions within Guangdong province (GDo), and neighboring provinces (Neighbor) from various periods. (Unit: ppb. The colorbar range of each sub-figure is same as the one in Figure 6)

3.4 Verification of the TSA by comparing to Zero-out Experiments

Here, the emission zero-out sensitivity experiments, another commonly used source apportionment method, were also conducted to evaluate the result from the TSA method. The zero-out method need to conduct two sets of simulation including the control run and zero-out run. In the control run, the simulations were conducted using the complete emissions. In the zero-out runs, the simulations were conducted with the emissions that specific period and area were removed. After that, the contribution of the specific source area and source time was derived by calculating the difference between the control and zero-out simulations. For each target date, the emission control area was set as the GBA, Guangdong province (GD), Guangdong and neighboring provinces (GD_Neighbor), respectively. The emission control period was set as continuous control beginning from the current day, which is also the target day (Day-0), from 1day ago (Day-1), from 2days ago (Day-2) and from 3days ago (Day-3), respectively. The zero-out experiments were carried out for the periods between 7th and 10th July (Typhoon case) and between 24th and 26th (Sub-tropical high case). More configurations can be found in Tables S4- S5.

From the result of zero-out experiments (Fig.9 and Fig.S9), it can be seen that, for the typhoon case (Fig.9), when only controlling the emission within the GBA, there is little difference between results of controlling emission 1 day in advance and 3 days in advance. This is consistent with the TSA result that the influence of the emission within the GBA is usually limited to 2 days. The effect of controlling emission 1 day ahead in GD is better than that of only GBA. There is less variation of the O_3 concentration when controlling the emission within Guangdong province 2 days and 3 days in advance. Meanwhile, regarding only emission control from the Day-0, it shows limited improvement in controlling the emission for a larger area (GD and GD_neigh) than solely within GBA. It's the same as the TSA result that the pollutants from neighboring provinces took effect on the O_3 over the GBA region at least 1 day later. Joint control from Guangdong and neigboring province (GD_neighbor) has a larger optimal effect in the Day-2 to Day-0 and Day-3 to Day-0 simulations. And the difference between GD_neighbor and the GD result is also more obvious in these simulations, indicating that it's more effective to implement joint control within other provinces 2-3 days in advance.

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For the sub-tropical high case (Fig.S9), whatever controlling the emission in the current day or 2 days ahead, the effect of only controlling emission within GBA is similar to that of joint controlling larger area (GD and GD_neigh). It supports our previous conclusion that the pollution is mainly contributed by the local source. At the same time, there is limited optimization effect to control the emission 2-3 days in advance than controlling 1 days in advance. To alleviate this ozone pollution, controlling the local emission within short-term should be effective. Although the contribution discrepancies between the source contribution (%) calculated from the zero-out method and the one from the TSA method can reach 20%, which is due to the non-linear chemistry relationship between ozone and its precursors and the mechanism of different methods (Kwok et al., 2015; Clappier et al., 2017), similar relationships between source area/time and receptor can be drawn. These results also support the validity of the TSA approach.

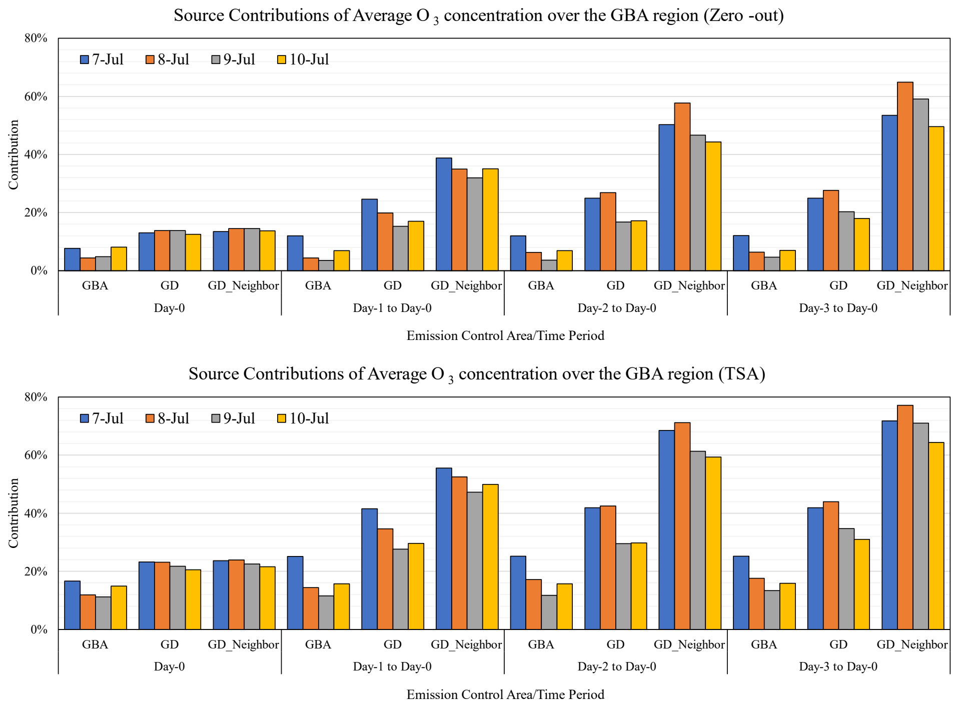

Figure 9. The contribution of different source areas and time periods to the O_3 concentration over the GBA in the typhoon case using the zero-out and TSA methods. (Different colors represent different target dates; Upper: Zero-out; Bottom: TSA)

3.5 Discussion

Previous studies mainly focused on exploring the contribution and control of various source areas and categories on O_3 over the GBA. The analysis in this study illustrated that there could be a larger difference between the temporal contribution of emissions to the O_3 pollution over the GBA under different weather patterns. It indicates that understanding the contribution of pollutants from different emitting periods and finding out the major period is also crucial in control policymaking, especially in episodic cases. Different from the zero-out method that needs sets of simulations, it can provide an overall picture of source contributions within one simulation, saving more computation costs, and is suitable for applications in which more potential sources are considered.

In addition, meteorological conditions play an important role in affecting the effectiveness of the emission control area and period. The results here suggest that the approach of typhoons usually strengthens the cross-region transportation of pollutants to the GBA. Therefore, cross-province collaboration and control should be

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implemented at least 2-3 days ahead when the typhoon is predicted. In contrast, local emission control within 2 days is more effective when the GBA is under the influence of a high-pressure system. Our findings emphasize the importance of considering the impact of meteorological conditions when implementing control measures in advance. The TSA method could help to better understand the spatial-temporal sources of O_3 pollution under the current emission level. It will provide scientific references for designing of effective and timely control policies when unfavorable weather conditions are predicted. So, the spatial-temporal influence of emission to O_3 over the GBA under other unfavourable conditions and seasons is also essential to further explore through the TSA method, which help to gain a more comprehensive understanding of when and where the O_3 over the GBA comes from.

Under the background of climate change, extreme weather, such as extreme heatwaves (Coffel et al., 2018; Dong et al.,2023), may occur more frequently, which will largely impact the source and sink of pollutants by different physical and chemical processes. At the same time, various emission control strategies responding to climate change, such as carbon neutrality (Liu et al., 2021; Zhang et al, 2021), will be implemented by governments in different countries, which will also change the structure of emissions. How these extreme weather and control measures influence the temporal characterization of sources and formation of air pollution, as well as the spatial-temporal contribution of emissions from different countries and their interactions, are also worth further investigation in the future. It can promote the mutual cooperation among nations to combat the environmental issues together.

However, it should be noted that the numerical model source apportionment results are usually influenced by the uncertainties of the emission inventory as most of the emission inventories are built up by the bottom-up method and cannot be updated in a timely manner. With the increasing availability of different types of observations, including surface monitoring and satellite remote sensing data, different top-down methods such as data assimilation (East et al., 2022) and machine learning (Chen et al., 2023) have been applied to integrate observations and optimize the emission. These methods should be implemented to update the emission inventory and combined with the TSA method to evaluate the evolution of spatial-temporal sources in different historical periods and provide up-to-date source information for policymaking.

4. Conclusion

In this study, we applied the CAMx-TSA method to analyze the spatial and temporal contribution of different sources to the O_3 pollution in the GBA in summer. The result shows that the O_3 over the GBA in summer is mainly contributed by the pollutants from local emissions, followed by other regions within Guangdong province (GDo) and neighbouring provinces. The O_3 was usually formed by the pollutants emitted within 3days, which account for more than 70%. During the O_3 episodes, when the typhoon moved from the eastern Philippine Sea to southern China, the prevailing wind shifted from south to north over the GBA. It conducive more pollutants transported from GDo and neigbouring provinces to the GBA, leading to an increase in O_3 concentrations. The pollutants emitted 3 days ago still have a significant contribution. While the typhoon just stayed near the sea areas east of Taiwan province and moved northly, under the continuous influence of northly wind, the emission of eastern China, even the North China Plain from 3 days ago can also have an obvious impact on O_3 over the GBA. In contrast, when the GBA is mainly under the control of sub-tropical high-pressure system, the ozone pollution was mainly caused by the local pollutants within the current 2 days. The results indicated that joint emission control action with other provinces 2-3 days in advance is more effective for preventing the O_3 pollution in the GBA when the typhoon is moving towards southern China. On the other hand, it's more efficient to pay more attention to local sources control within 2 days when the GBA is under the control of the high-pressure system.

Here, different surrounding provinces were categorized as one source area here to save computation resource for more potential source investigation. As the neigbouring province was illustrated as a major contributor to the O_3 in the GBA, it is necessary to further divided this source into several sub-source areas and explore their individual impact in future work. In addition, individual source categories were not separated in this study, mainly due to the application of different emission inventories with different source category classifications, making it difficult to combine them. It is important to note that each source category has its own characteristic temporal profile, which can have different temporal impacts on O_3 concentrations. Therefore, the temporal contribution of various source categories, including anthropogenic and biogenic emissions, should be also considered in future work. These works can provide more spatial and temporal information of O_3 source over the GBA to the local governments so that the targeted control measures can be designed and implemented more effectively and timely.

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Code and Data availability

Hourly O_3 observation data were released by the China National Environmental Monitoring Centre (http://www.cnemc.cn/en, last access 24 December; CNEMC, 2023) and the Hong Kong Environmental Protection Department (https://cd.epic.epd.gov.hk/EPICDI/air/station/?lang=en, last access 24 December 2023; HKEPD, 2023). The CAMx model code is freely available via https://www.camx.com/download/, last access 24 December, 2023)

Author contribution

CY, LX, and JF designed the research. CY contributed to model development, simulation and data analysis. LX and JF contributed to the result discussion. CY prepared the manuscript with contributions from all co-authors.

Competing interests

The authors declare that they have no conflict of interest.

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