Source apportionment of ozone under different synoptic patterns in the Pearl River Delta region

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1. INTRODUCTION

In recent years, with the effective control of fine particulate matter ($PM_{2.5}$) by the government, the concentration of $PM_{2.5}$ in the Pearl River Delta (PRD) region has decreased gradually (Lin et al., 2018). However, the pollution problem characterized by a high concentration of ozone is sequentially emerging (Yang et al., 2020, State of the Environment in China, 2019). In the case of relatively stable emissions, the meteorological condition is an essential factor affecting ozone pollution (Liu et al., 2020). Therefore, exploring the contribution of various source regions and source categories to the O_3 concentration under different synoptic patterns is an integral part of regional atmospheric environmental research.

2. METHOD

In this work, the Weather Research Forecast (WRF)-Sparse Matrix Operator Kernel Emission (SMOKE) Comprehensive Air Quality Model with extension (CAMx) modeling system with Ozone Source Apportionment Technology (OSAT) module was applied to analyze the influence of particular synoptic patterns. The MEIC emission inventory for 2012 was applied for the regions outside the PRD in D1 and D2. A highly resolved emission inventory for 2015 provided by the Hong Kong Environmental Protection Department (HKEPD). Three nested domains with resolutions of 27km, 9km, and 3km were set for the simulation. The 10 major cities shown in Fig. 1(b) are separated, and the tracked emission sources are divided into six categories, namely area, mobile, industrial, power plant, biogenic, and marine-ship emissions. The above-mentioned simulation results were used to analysis the ozone maximum daily 8-hour average (O_3) in the 10 major cities in the PRD region under three synoptic patterns. Three episodic cases were chosen under the three different synoptic patterns, namely sea high pressure, equalizing pressure field and subtropical high pressure.

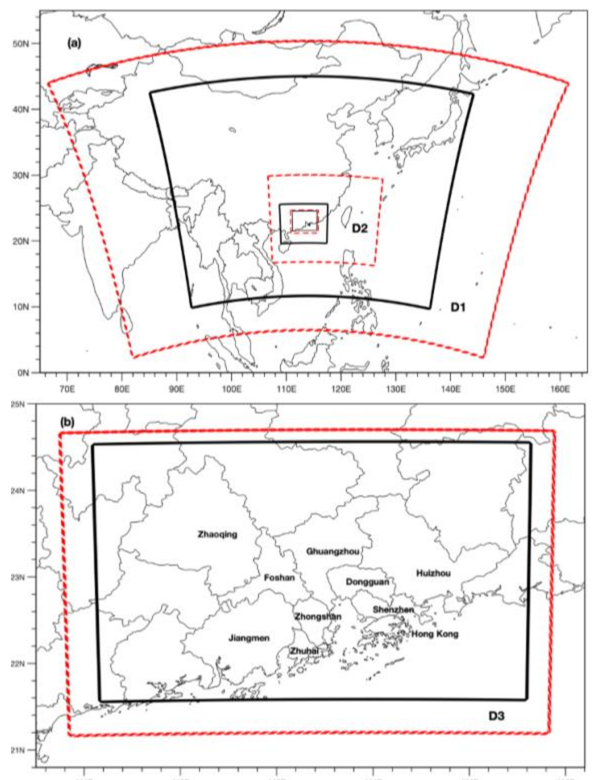

Fig. 1. Domain extents for WRF (red dashed lines) and CAMx (solid black lines), including (a) Domain1 (D1), Domain2 (D2), and (b) Domain3 (D3). Red dots represent the measurement sites. 10 major cities in the PRD region are labeled on the map.

3. RESULTS AND DISCUSSIONS

3.1 Monthly Ozone Concentration and Source Apportionment

As shown in Fig. 2, in summer, the solar radiation is strong, which is conducive to the formation of O_3, so the monthly average

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concentration of ozone is the highest in the August. Foshan, Dongguan, Guangzhou is the most serious area of ozone pollution in summer.

In January and October, when northerly wind prevails, the O_3 concentration's contribution was mainly influenced by the emissions outside the PRD region, and the contribution can reach more than 80%. For April and August, under the control of weak southerly wind, the emissions contribution outside the PRD region was reduced to approximately 60-70%.

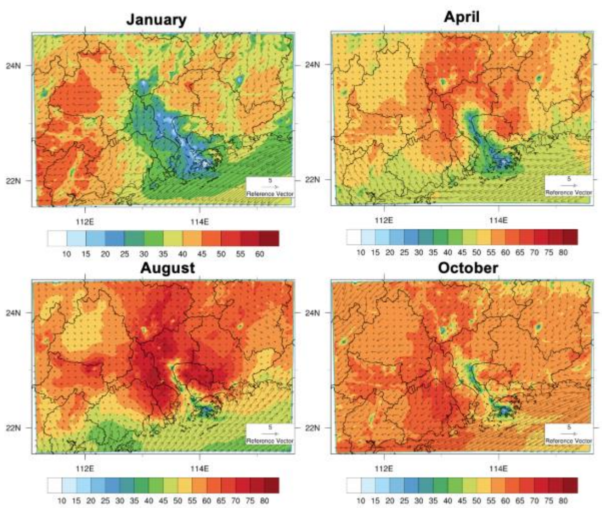

Fig. 2. Monthly O_3-8h concentration spatial distributions with wind fields in January, April, August, and October over the PRD region (Units: ppb)

Monthly source contribution in percentage(%) are displayed in Fig. 3. In the four seasons, mobile, area, and biogenic sources were the three main sources of ozone. For cities with more motor vehicle ownerships, such as Guangzhou and Shenzhen, the mobile contribution can be higher than 40%. For Hong Kong, the contribution of ship emissions cannot be neglected. The biogenic emission is an important source for Huizhou. Although the contribution of several sources varied slightly in different months, the overall change was not significant. In the next step, it's necessary to analyze the characteristics of ozone concentration, source categories, and source regional contributions under different meteorological conditions.

Fig. 3. Monthly source contributions in percentage(%) to O3-8h in January, April, August, and October in the PRD region

3.2 Concentration and Source Category Contribution

The average O_3 concentration under three synoptics patterns are compared with the four-month average concentrations of different cities in the PRD region as shown in Fig. 4. It is obvious that the concentration of ozone got increased under three meteorological conditions. The ozone concentration in most cities except Hong Kong and Shenzhen are larger than World Health Organization (WHO) Air Quality Guideline (AQG), which is 100 μg/m^3. The concentration in Foshan can reach around 160 μg/m^3 under the equalizing pressure field. The ozone concentration increases most under the equalizing pressure field compared with the four-month average concentration, with a magnitude increase of 26%. The magnitudes in the sea high pressure and subtropical high pressure increased by around 8% and 11%.

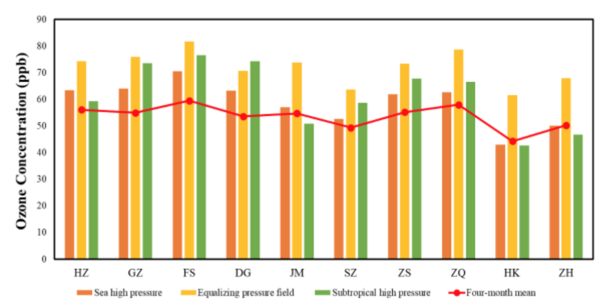

Fig. 4. The average O3-8h concentrations under three synoptics patterns and four-month average concentrations of different cities in the PRD region (Units: ppb)

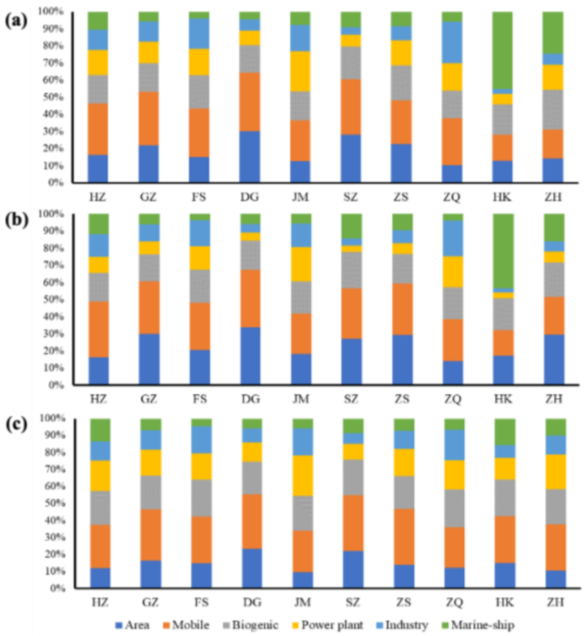

Fig. 5. Source categories' contributions to O_3 in the PRD region under (a) sea high pressure, (b) equalizing pressure field, and (c) subtropical high pressure.

Among these 10 major cities in the PRD region, Guangzhou, Dongguan, and Foshan were most affected by meteorological conditions. The source contribution of ozone in these synoptic patterns are different from the general situation. Among the six sources in the Pearl River Delta, the contribution of mobile sources is outstanding under all three weather conditions. After the mobile emissions, biogenic sources and area sources attribute to the highest contribution under subtropical high pressure and sea high pressure, respectively. Different weather conditions have a slight effect on source categories. It is indicated that the source categories are more closely related to the emission and are less affected by the weather conditions. Therefore, it is necessary to analyze source regional contribution in the episodic cases.

3.3 Local, Regional and Superregional contribution

In this study, three types of source regions are defined. Local sources represent the sources from a local city. Regional transport means the sources from another city within the PRD region. Superregional transport is the emissions outside the PRD region. Variations in local, regional, superregional ozone contribution compared with monthly contributions are shown in the right Table. High concentration represents ozone daily maximum 8-hour average concentration. O_3 had more obvious changes in the high concentration period under different synoptic patterns. As the equalizing pressure field changed the direction of the southerly prevailing wind in April, the contribution of superregional transport increased by 6.1%, compared with the monthly contribution. With the effect of the subtropical high pressure, the local contribution increased significantly, especially in Jiangmen (+8.8%) and Shenzhen (+7.3%), compared with the monthly mean contribution.

Table. 2. Variations in local, regional, superregional O_3 contribution compared with monthly contributions in 10 cities over the PRD region under sea high pressure, equalizing pressure, and subtropical high pressure synoptic patterns. High conc and low conc represent O_3 daily maximum 8-hour average concentration and other hour average concentration.

			HZ	GZ	FS	DG	JM	SZ	ZS	ZQ	HK	ZH
Sea high pressure	High conc.	Local	1.8%	4.3%	5.7%	4.2%	4.6%	5.3%	3.0%	2.6%	1.7%	2.8%
		Regional	7.6%	7.7%	4.6%	8.8%	-0.5%	1.8%	3.4%	3.2%	-2.2%	-5.3%
		S-Regional	-9.4%	-12.0%	-10.2%	-13.0%	-4.1%	-7.2%	-6.4%	-5.8%	0.5%	2.5%
	Low conc.	Local	0.7%	1.2%	0.6%	1.0%	0.6%	0.6%	0.3%	0.1%	0.6%	0.4%
		Regional	5.0%	5.3%	6.1%	5.8%	3.7%	3.7%	3.9%	4.0%	3.6%	3.7%
		S-Regional	-5.7%	-6.5%	-6.8%	-6.8%	-4.3%	-4.2%	-4.2%	-4.2%	-4.2%	-4.1%
Equalizing pressure field	High conc.	Local	-1.7%	-2.8%	-1.8%	-3.7%	-3.0%	-3.9%	-1.6%	-1.7%	-0.6%	-1.5%
		Regional	-4.8%	-0.8%	-2.8%	-4.7%	-6.6%	-3.8%	-8.0%	-2.2%	-1.0%	-4.2%
		S-Regional	6.5%	3.6%	4.6%	8.4%	9.6%	7.7%	9.6%	4.0%	1.6%	5.7%
	Low conc.	Local	-0.5%	-0.3%	-0.1%	-0.2%	-0.3%	-0.2%	0.0%	-0.1%	-0.2%	0.0%
		Regional	-1.7%	-1.0%	-1.2%	-1.1%	-2.3%	-2.3%	-2.3%	-2.2%	-1.9%	-1.9%
		S-Regional	2.1%	1.3%	1.3%	1.3%	2.6%	2.5%	2.3%	2.4%	2.1%	2.0%

Sub-tropical high pressure	High conc.	Local	7.0%	5.9%	6.0%	6.3%	8.8%	7.3%	5.5%	2.1%	2.4%	4.7%
		Regional	5.9%	11.3%	10.0%	11.2%	5.3%	9.3%	9.2%	16.1%	12.8%	7.8%
		S-Regional	-12.8%	-17.2%	-16.0%	-17.5%	-14.1%	-16.7%	-14.7%	-18.2%	-15.2%	-12.6%
	Low conc.	Local	5.8%	2.5%	1.1%	2.2%	2.1%	2.3%	0.6%	0.4%	1.6%	1.0%
		Regional	9.1%	14.5%	16.2%	15.5%	12.0%	11.9%	13.7%	13.7%	12.8%	13.5%
		S-Regional	-14.9%	-17.0%	-17.3%	-17.7%	-14.0%	-14.2%	-14.3%	-14.1%	-14.4%	-14.5%

3.4 Regional Contributions to Guangzhou and Shenzhen

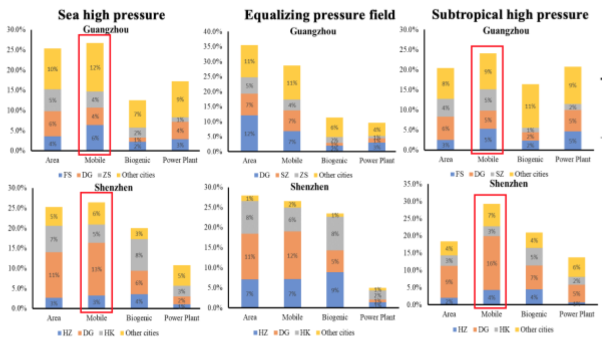

Fig. 6. Regional categories' contributions to Guangzhou and Shenzhen cities in the PRD region under different synoptic patterns. The x-axis is the top 4 regional source categories' contributions. The colored bars represent the top three contributors in neighboring cities for the specific source category. The yellow bar represents the contribution from the rest of the cities.

Table. 3. Regional contributions of O3-8h from surrounding cities to Guangzhou (GZ) and Shenzhen (SZ) in the PRD region.

	Sea high pressure		**Equalizing pressure field**		**Subtropical high pressure**	
	GZ	SZ	GZ	SZ	GZ	SZ
HZ	7.1%	12.1%	0.1%	28.2%	0.2%	13.3%
GZ	-	2.9%	-	0.9%	-	10.5%
FS	17.8%	1.9%	10.9%	0.1%	17.8%	1.9%
DG	20.0%	33.7%	27.4%	30.4%	22.3%	40.9%
JM	13.4%	4.4%	5.4%	1.1%	12.6%	3.5%
SZ	11.3%	-	17.4%	-	13.4%	-
ZS	14.1%	4.5%	12.8%	1.0%	9.0%	3.6%
ZQ	1.1%	0.2%	0.1%	0.0%	1.8%	0.4%
HK	6.4%	32.6%	10.2%	36.3%	8.1%	20.7%
ZH	8.7%	7.8%	6.3%	2.1%	6.8%	5.2%

Guangzhou and Shenzhen are two critical cities with dense population and mature urbanization scale in the Pearl River Delta region. Also, these two cities are most affected by regional sources from surrounding cities. Dongguan is an important regional source of Guangzhou and Shenzhen under three synoptic patterns. Under the control of the subtropical high pressure, the contributions from Dongguan can reach 22% and 40% in Guangzhou and Shenzhen. Mobile source accounts for more than 20% for the regional transported O_3 under the sea high pressure and subtropical high pressure in both Guangzhou and Shenzhen. The contribution of area source is the most significant in equalizing pressure case, which can be up to 25%.

3.5 Diurnal variation in Regional contributions

Because ozone concentration is affected by solar radiation, its diurnal variation is apparent. It is imperative to study the diurnal variation of ozone under three different episodic cases to implement effective ozone control policies. In the three synoptic patterns, the diurnal variation of O_3 concentration had similar characteristics with a single peak with the maximum in the daytime. At 6:00-8:00 a.m., a large amount of nitric oxide emitted by motor vehicles as a reducing agent. Then nitric oxide reacts with ozone during the traffic peak, resulting in a significant decrease in the concentration of O_3. Then in the presence of sufficient sunlight (UV radiation), O_3 is formed from the complex photochemical reactions of primary emitted NOx and VOC.

In the equalizing pressure field, the contribution from Shenzhen increased sharply from 5:00 a.m. to 8:00 a.m. The atmospheric stratification near the surface was relatively stable, leading to the rapid accumulation of O_3 precursors. Under the control of subtropical high pressure, the contribution of Huizhou in nighttime was significantly reduced. In contrast, Hong Kong and Dongguan's regional contributions increased, which indicates that the weather condition changes the long-range transport of surface O_3 at night.

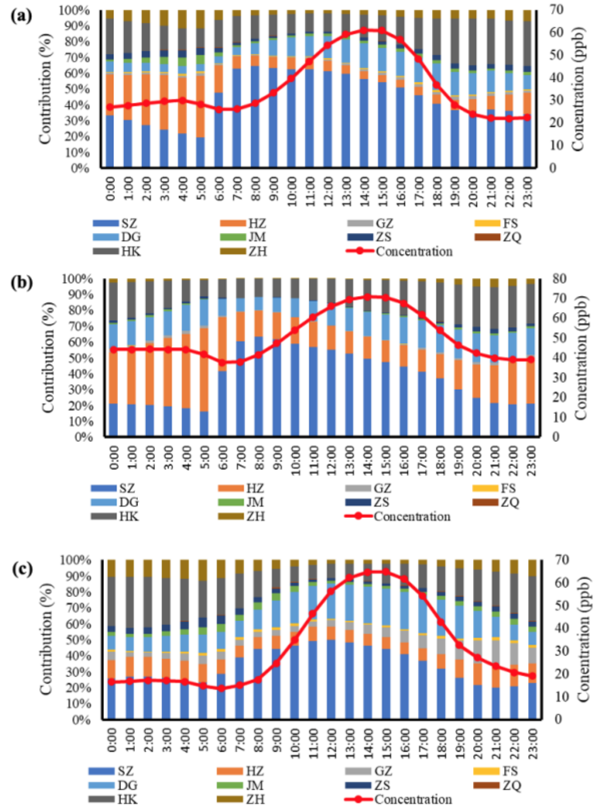

Fig. 7. Diurnal variation of O_3 concentrations (red lines, units: ppb) and regional contributions to O_3 in Shenzhen city under (a) sea high pressure, (b) equalizing pressure field, and (c) subtropical high pressure.

4. CONCLUSIONS

In conclusion, the concentration of ozone increased under three meteorological conditions. Under distinctive synoptic patterns, the emissions outside PRD have the most significant contribution invariably. Among the six sources in the Pearl River Delta, the contribution of mobile sources is outstanding under all three weather conditions. After the mobile emissions, biogenic sources and area sources attribute to the highest contribution under subtropical high pressure and sea high pressure, respectively. In the equalizing pressure field, the contribution of superregional transport increased by 6.1%. With the effect of the subtropical high pressure, the local contribution increased significantly, especially in Jiangmen (+8.8%) and Shenzhen (+7.3%). Our results indicated that collaborative emission control measures should be strengthened with the surrounding area. Combined with the meteorological situation, controlling the endogenous emission in the PRD plays a pivotal role in preventing O3 pollution.

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