Characteristics of Secondary PM$_{2.5}$ Under Different Photochemical Reactivity Backgrounds in the Pearl River Delta Region

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With the increasing control of air pollution, the levels of atmospheric particulates in the Pearl River Delta (PRD) region are gradually decreasing. However, ozone pollution has become more and more serious, and the problem of secondary aerosol pollution caused by photochemical reactions cannot be ignored. Based on the observation data of environmental and meteorological stations in the nine cities of the PRD during 2019, we investigated the variations of secondary PM$_{2.5}$ (PM$_{2.5}$-sec) in the PRD under different photochemical reactivity backgrounds. It was shown that the photochemical reactivities appeared more significant in the central and western areas than those in the eastern areas of the PRD and appeared more significant in inland areas than those in coastal areas. The days of moderate and high photochemical reactivities mainly appeared from August to November. PM$_{2.5}$-sec concentrations were the highest in autumn, during which the regional discrepancies appeared most significantly with the highest levels in the southern areas. With the enhancement of the photochemical reactivity background, the PM$_{2.5}$-sec level at each station increased significantly, which appeared significantly higher in coastal areas than in inland areas. Both PM$_{2.5}$-sec and ozone concentrations showed single-peak variations, which appeared higher in the daytime than at night with the peak occurring at about 15:00. For each pollutant, the average maximum concentration appeared higher for polluted stations than for clean ones, indicating that the atmospheric oxidation background was conducive to the formation of PM$_{2.5}$-sec.

Keywords: secondary PM$_{2.5}$, ozone, photochemical reactivity, oxidation background, coordinated increase

INTRODUCTION

With the strengthening of environmental governance, air pollution in some cities of China has been gradually transforming from single to complex pollution as a result of the increase in ozone and aerosol levels (Zhang et al., 2019; Wang et al., 2021a; Wang et al., 2021b). Ozone and aerosols significantly influence human health, crop yield, and global climate change (Mills et al., 2009; Stoker et al., 2013; Zhang et al., 2017; Fleming et al., 2018), meaning that their coordinated control is very urgent. Therefore, many scholars have carried out relevant studies and found that their coordinated variations are fulfilled through the influence of aerosols on the levels of atmospheric radiation and hydroxyl radicals and...
the influence of ozone on atmospheric photochemical reactivities (Yuan et al., 2012; Liu et al., 2021; Wang et al., 2021a; Wang et al., 2021b; Wang et al., 2021c). It is worth noting that the strengthening of atmospheric photochemical reactivities and the following significant growth of secondary aerosols resulting from ozone enhancement have become the common causes of complex pollution in China. However, because of the influence of weather background, emission source structure, and geographical factors, the characteristics of complex pollution are different (Liu et al., 2019; Zhao et al., 2021). Some scholars found that with the increase of PM2.5 levels, the maximum values and change rates of ozone concentrations with different backgrounds in Guangzhou increase gradually, indicating the coordinated increase between PM2.5 and ozone (Yao et al., 2021). Some scholars found that in Handan, the fourth largest city of Hebei Province, ozone increased with the enhancement of PM2.5 during summer in the case of its concentration being below the standard values, whereas high-level PM2.5 had an inhibitory effect on ozone formation in winter (Zhao et al., 2021).

The common precursors of aerosols and ozone (i.e., VOCs and NOx) can be transformed into secondary aerosols and inorganic salts through gas–particulate matter interactions (Xing et al., 2017), contributing to the formation of PM2.5. Li (Li et al., 2020) evaluated the coordinated increase of ozone and secondary aerosols in Beijing, Shanghai, and Guangzhou and found that the formation of secondary aerosols in Shanghai and Guangzhou increased by multiples with the enhancement of photochemical reactivities, whereas that in Beijing was scarcely varied. Previous studies were mainly based on a single city or station and rarely focused on regional ozone and secondary aerosol variations (Wu et al., 2021).

The Pearl River Delta (PRD) region is located south of the Tropic of Cancer, bordering the South China Sea, with good thermal conditions. The regional economy is developed, the manufacturing industry is prosperous, and the population is dense. The precursors produced by a large number of industrial sources and man-made sources are conducive to the generation of ozone photochemical reactions. In recent years, the ozone levels in the PRD have gradually increased (Zhan, 2018; Yin and Wang, 2020), with complex pollution of ozone and secondary aerosols occurring frequently (Lai et al., 2018). In this article, we analyze the various characteristics of PM2.5, secondary PM2.5 mass concentration in the PRD under different photochemical reactivity backgrounds, based on the observed data from environmental and meteorological stations in nine cities of the PRD in 2019; the correlation between ozone pollution and secondary PM2.5 were revealed; and the possible mechanisms of formation of PM2.5, secondary PM2.5 under different ozone levels were analyzed, which provide scientific support for the coordinated control of PM2.5 and ozone in the PRD.

**DATA AND METHODS**

**Data Sources**

This article selects the PRD as the research area including nine cities of Guangdong Province, i.e., Zhaoqing, Guangzhou, Foshan, Zhuhai, Jiangmen, Zhongshan, Huizhou, Shenzhen, and Dongguan. The data come from the national control station of the Department of Ecology and Environment of Guangdong Province (Station No. 1345A-1400A, **Figure 1**). Season classification criteria are as follows: spring (March, April, and May), summer (June, July, and August), autumn (September, October, and November), and winter (December, January, and February). The validities of atmospheric pollutant concentration data meet the requirements of the Environmental Air Quality Standard (GB3095-2012) and the Technical Specification for Environmental Air Quality Assessment (TRIAL) (HJ633-2013). The meteorological data come from the national basic meteorological observation stations of the respective cities, and the accuracy of all meteorological data was over 98% after quality control. Data on the 10 m mean wind and accumulated rainfall in the PRD in 2019 were obtained from the EC ERA5 monthly reanalysis data.

**Research Methods**

Li et al. (2020) used the following research methods to analyze the coordinated increase of ozone and secondary aerosols in typical Chinese cities: First, the daily maximum 1 h average concentration of ozone (O3-max) was used to classify the atmospheric photochemical reactivity level. O3-max < 100 μg/m³ was defined as a low level of photochemical reactivity (O3L), and 100 μg/m³ ≤ O3-max < 160 μg/m³ was defined as a light level of photochemical reactivity (O3LH). 160 μg/m³ ≤ O3-max < 200 μg/m³ was defined as a moderate level of photochemical reactivity (O3M), and O3-max ≥ 200 μg/m³ was defined as a high level of photochemical reactivity (O3H). Then, CO was used as a tracer of the primary emission source, assuming that the structure of the emission source is basically stable; the larger the ratio of PM2.5/CO mass concentration is, the larger the proportion of secondary components in PM2.5 is (Chang and Lee, 2007; Zhang et al., 2015). Specific equations are as follows:

\[
(PM_{2.5})_{p,i,t} = CO_{i,t} \times (PM_{2.5}/CO)_{p,t},
\]

**FIGURE 1 | Distribution of national control stations in the Pearl River Delta.**
The coordinated increase of PM2.5 and ozone. In this article, the reactivities. Finally, since ozone concentrations in eastern China aerosol for the calculation of different levels of photochemical concentrations increasing simultaneously and generally peak between 14:00 and 16:00, the phenomenon of PM2.5 and CO LH respectively. CO LH is observed PM2.5 concentration. t is the specific time of the day. L, LH, M, and H represent O3L, O3LH, O3M, and O3H, respectively. CO LH × (PM2.5/CO) LH is 25% of the hourly PM2.5/CO in O3H. It is the reference value of the primary aerosol for the calculation of different levels of photochemical reactivities. Finally, since ozone concentrations in eastern China generally peak between 14:00 and 16:00, the phenomenon of PM2.5 and ozone concentration increasing simultaneously and continuously for no less than 2 h from 11:00 to 19:00 is defined as the coordinated increase of PM2.5 and ozone. In this article, the aforementioned methods are used to estimate the hourly concentration of PM2.5-sec under different photochemical reactivities and to calculate the time for the coordinated increase of PM2.5-sec and ozone. More details can be found in Li et al. (2020).

**DISTRIBUTION CHARACTERISTICS OF AIR POLLUTANT CONCENTRATION IN THE PEARL RIVER DELTA**

**Annual Average Variations**

Figure 2 shows the mean concentrations of PM2.5, NO2, O3-8h, and CO at various stations in the PRD in 2019. The air pollutants have obvious spatial variation characteristics. Concentrations of PM2.5, NO2, and CO are higher in the central and northern parts of the PRD (e.g., Guangzhou, Foshan, and Dongguan), whereas they are lower in coastal areas. The maximum annual mean concentrations of PM2.5, NO2, and CO appear in Dongguan (37 µg/m³), Guangzhou (54 µg/m³), and Foshan (980 µg/m³), respectively. The average ozone concentrations in coastal areas (e.g., Zhuhai, Shenzhen, and Huizhou) are higher than those in other areas, and the maximum appears in Shenzhen with a value of 72 µg/m³.

**Seasonal Variations**

Table 1 shows the seasonal variations of air pollutants in the PRD in 2019. The average concentrations of PM2.5, CO, and NO2 in the PRD appear the highest in winter and the lowest in summer, whereas the concentrations of O3-8h are highest in autumn and the lowest in winter. The concentrations of PM2.5, NO2, and CO in the northern part of the PRD are higher than those in the southern part, whereas the distributions of O3-8h appear vice versa. Figure 3 shows the variations of PM2.5 and ozone with temperature and humidity during autumn and winter in the PRD. The high-level PM2.5 in autumn mainly occurs in the environment with humidity over 80%, but there is no similar distribution in winter. High ozone concentrations in autumn and winter appear under high-temperature and low-humidity conditions. According to the 10 m average wind field and cumulative rainfall distribution in the PRD (Figure 4), the prevailing wind directions in spring and summer are southeast and southwest, which bring clean marine air mass. Meanwhile, abundant precipitation is also conducive to pollutant removal. In autumn and winter, the prevailing wind changes to the northeastern direction, leading to inland polluted air masses influencing the air quality of the PRD, and the precipitation decreasing significantly compared with that in
TABLE 1 | Seasonal variations of air pollutants in the PRD.

| City     | Pollutant | Spring | Summer | Autumn | Winter | Average |
|----------|-----------|--------|--------|--------|--------|---------|
| Dongguan | PM$_{2.5}$ | 26.6   | 18.7   | 40.7   | 42.8   | 32.2    |
|          | NO$_2$    | 35.9   | 28.4   | 39.5   | 43.3   | 36.8    |
|          | O$_3$-n   | 49.4   | 60.8   | 89.1   | 44.9   | 61.0    |
|          | CO        | 0.767  | 0.699  | 0.781  | 0.92   | 0.789   |
|          | PM$_{2.5}$ | 25.6   | 17.6   | 37.8   | 39.1   | 30      |
| Foshan   | NO$_2$    | 41.9   | 28.5   | 44.3   | 49.9   | 41.2    |
|          | O$_3$-n   | 38.2   | 53.1   | 82.8   | 34.7   | 52.2    |
|          | CO        | 0.837  | 0.733  | 0.813  | 0.813  | 0.857   |
| Guangzhou| PM$_{2.5}$ | 24     | 18.1   | 37     | 38.8   | 29.5    |
|          | NO$_2$    | 44     | 32     | 45     | 50.1   | 42.8    |
|          | O$_3$-n   | 39.4   | 51.9   | 79     | 37.4   | 51.9    |
|          | CO        | 0.794  | 0.664  | 0.795  | 1.013  | 0.817   |
| Huizhou  | PM$_{2.5}$ | 20.5   | 14.4   | 31.3   | 33.8   | 25      |
|          | NO$_2$    | 22.7   | 19.9   | 26.5   | 29.1   | 24.6    |
|          | O$_3$-n   | 60.9   | 50.9   | 84     | 50.6   | 61.6    |
|          | CO        | 0.636  | 0.563  | 0.672  | 0.855  | 0.682   |
| Jiangmen | PM$_{2.5}$ | 22.8   | 15.4   | 34.6   | 36     | 27.2    |
|          | NO$_2$    | 28.6   | 17.4   | 36.7   | 46.5   | 32.3    |
|          | O$_3$-n   | 43.6   | 53.6   | 98.2   | 41.4   | 59.2    |
|          | CO        | 0.744  | 0.6    | 0.781  | 1.033  | 0.79    |
| Shenzhen | PM$_{2.5}$ | 19.7   | 13.2   | 31.7   | 32.3   | 24.2    |
|          | NO$_2$    | 22.9   | 20.1   | 26.5   | 32.4   | 25.5    |
|          | O$_3$-n   | 60.5   | 49.7   | 90.7   | 51.6   | 63.1    |
|          | CO        | 0.599  | 0.545  | 0.625  | 0.743  | 0.628   |
| Zhuhai   | PM$_{2.5}$ | 19.2   | 11.9   | 31.6   | 35.4   | 24.5    |
|          | NO$_2$    | 27.6   | 15.4   | 25.1   | 42     | 27.5    |
|          | O$_3$-n   | 55.5   | 52     | 96     | 51.3   | 63.7    |
|          | CO        | 0.67   | 0.541  | 0.817  | 0.86   | 0.672   |
| Zhaoqing | PM$_{2.5}$ | 29.8   | 19     | 35.6   | 41.3   | 31.4    |
|          | NO$_2$    | 32.5   | 21.2   | 34.2   | 39     | 31.7    |
|          | O$_3$-n   | 38.8   | 53.1   | 83     | 40     | 53.7    |
|          | CO        | 0.829  | 0.66   | 0.8    | 0.963  | 0.813   |
| Zhongshan| PM$_{2.5}$ | 20.5   | 12.9   | 34.3   | 38.6   | 26.6    |
|          | NO$_2$    | 23.8   | 14.1   | 34.4   | 46.4   | 29.7    |
|          | O$_3$-n   | 46.5   | 55.4   | 91.9   | 38.1   | 58     |
|          | CO        | 0.656  | 0.558  | 0.881  | 0.895  | 0.698   |
|          | PM$_{2.5}$ | 23.2   | 15.7   | 35     | 37.6   | 27.9    |
|          | NO$_2$    | 31.1   | 21.9   | 34.7   | 42.1   | 32.4    |
|          | O$_3$-n   | 48.1   | 53.4   | 88.3   | 43.3   | 58.3    |
|          | CO        | 0.726  | 0.617  | 0.729  | 0.925  | 0.749   |

*Concentration of air pollutants: (1) PM$_{2.5}$, μg/m$^3$; (2) NO$_2$, μg/m$^3$; (3) O$_3$, μg/m$^3$; (4) CO, mg/m$^3$. 

Seasonal Variation of the Coordinated Increase of PM$_{2.5}$ and Ozone

Figure 7 shows the monthly averaged days of different photochemical reactivities and average hours of the coordinated increase in 2019. O$_{3M}$ and O$_{3H}$ mainly concentrate from August to November, among which the maximum O$_{3M}$ days appear in November (10.1 days), whereas the maximum O$_{3H}$ days appear in September (11.7 days). What is more, the trend of coordinated increase time is basically opposite to that of O$_{3L}$ days. Therefore, a coordinated increase of PM$_{2.5}$ and ozone mainly occurs under more significant oxidation backgrounds.

In general, photochemical reactivities in the PRD have obvious spatial and temporal variation differences. In terms of spatial variations, photochemical reactivities appear high in central and western regions but low in eastern regions and high in inland regions but low in coastal regions. In terms of seasonal variations, moderate and high levels of photochemical reactivities mainly...
appear in autumn. In general, higher levels of photochemical reactivities are more conducive to the coordinated increase of PM$_{2.5}$ and ozone, and the proportions of secondary aerosols in PM$_{2.5}$ are also higher. However, some coastal stations in Shenzhen and Zhuhai have a long-term coordinated increase or a high PM$_{2.5}$/CO value, which requires further research.

DISTRIBUTION CHARACTERISTICS OF PM$_{2.5}$-SEC UNDER DIFFERENT PHOTOCHEMICAL REACTIVITY BACKGROUNDS

Temporal and Spatial Variation Characteristics of PM$_{2.5}$-sec

Figure 8 shows the seasonal variation of the average PM$_{2.5}$-sec concentration at each station. The concentration in autumn is significantly higher than that in other seasons, and the concentration varies from 6 μg/m$^3$ to 13 μg/m$^3$, with the maximum appearing at Jida station in Zhuhai. PM$_{2.5}$-sec concentrations are the highest in autumn, during which the regional discrepancies appear the most significantly with the highest levels in the southern areas. High concentrations of PM$_{2.5}$-sec and ozone occur in the southern areas in autumn, indicating that the atmospheric oxidation background promotes the formation of PM$_{2.5}$-sec.

Variation Characteristics of PM$_{2.5}$-sec Under Different Photochemical Backgrounds

In addition to seasonal differences, there are also differences in PM$_{2.5}$-sec concentrations and the proportions of PM$_{2.5}$-sec in PM$_{2.5}$ under different photochemical reactivity backgrounds. Figure 9 shows the regional variations of PM$_{2.5}$-sec under different photochemical reactivities. With the enhancement of photochemical reactivities, PM$_{2.5}$-sec levels increase significantly. Under the background of O$_3$H, the annual average PM$_{2.5}$-sec concentrations at the coastal stations are generally above 13 μg/m$^3$ with a maximum of 20 μg/m$^3$ at some stations, whereas under the background of O$_3$M, the concentrations at the coastal stations are basically around 10 μg/m$^3$. Moreover, under the same photochemical reactivity background, PM$_{2.5}$-sec concentrations in coastal areas are significantly higher than those in inland areas. Figure 10 shows that the proportions of PM$_{2.5}$-sec

![Figure 3](image_url)
in PM$_{2.5}$ have similar characteristics under different photochemical reactivity backgrounds. The ratios are about 10% in O$_{3L}$, 20%–30% in O$_{3M}$, and 20–50% in O$_{3H}$.

Meanwhile, under the same photochemical background, the ratios of PM$_{2.5-sec}$/PM$_{2.5}$ in coastal areas are significantly higher than those in inland areas.
FIGURE 6 | Coordinated increase time and PM$_{2.5}$/CO distribution in the PRD.

FIGURE 7 | Monthly averaged days of different photochemical reactivities and the average hours of coordinated increase.

FIGURE 8 | Seasonal variations of PM$_{2.5}$-sec in the PRD.
Diurnal Variation Characteristics

Because the stations in the PRD are widespread with significant background discrepancies, we refer to the standard of ozone pollution in the Ambient Air Quality Standard (GB3095-2012) and take MDA8-O₃ (daily maximum 8 h average value of O₃) ≥ 160 μg/m³ as the classification standard. Then, the three stations with the most days exceeding the standard (located in Guangzhou, Foshan, and Dongguan) are identified as polluted stations, whereas the three stations with the least number (located in Zhuhai, Shenzhen, and Huizhou) are defined as clean stations.

Finally, diurnal variations and standard deviations of PM₂.₅-sec and ozone are presented (Figure 11, the error bars depict the standard deviation). The concentrations of the two pollutants are higher in the daytime than at night, and their maximum concentrations for both appear at about 15:00. As for ozone, the variation at polluted stations is larger than that at clean stations because the maximum concentration of ozone at polluted stations (91 μg/m³) is higher than that at clean stations (60 μg/m³) due to the photochemical reactions of a large number of precursors in the daytime, whereas the minimum concentration

![Figure 9](image1.png) **FIGURE 9** | PM₂.₅-sec variations under different photochemical backgrounds.

![Figure 10](image2.png) **FIGURE 10** | Variations of the ratios in PM₂.₅-sec/PM₂.₅ under different photochemical backgrounds.
of ozone in polluted stations is lower than that in clean stations due to the consumption of a large number of NO in polluted stations at night. Under the background of high-photochemical reactivities, the concentration of PM$_{2.5}$-sec at polluted stations is higher than that at clean stations from 12:00 to 20:00, which further reflects the promoting effect of the atmospheric oxidation background on PM$_{2.5}$-sec formation.

**CONCLUSION**

Based on the observation data of environmental and meteorological stations in the nine cities of the PRD during 2019, the variations of PM$_{2.5}$-sec and the coordinated increase of PM$_{2.5}$-sec and ozone under different photochemical reactivity backgrounds in the PRD are revealed.

The trend of coordinated increase time is basically opposite to that of O$_3$L days, meaning that the coordinated increase of PM$_{2.5}$ and ozone mainly occurs under more significant oxidation backgrounds. The photochemical reactivities appear more significant in the central and western areas than in the eastern areas of the PRD and appear more significant in inland areas than in coastal areas. The days of moderate- and high-photochemical reactivities mainly appear from August to November.

The PM$_{2.5}$-sec concentrations are the highest in autumn, during which the regional discrepancies appear the most significantly with the highest levels in the southern areas. With the enhancement of the photochemical reactivity background, the PM$_{2.5}$-sec level at each station increases significantly, which appears significantly higher in coastal areas than that in inland areas. The ratios of PM$_{2.5}$-sec/PM$_{2.5}$ are as high as 30–50% under the high-photochemical reactivity background.

Both PM$_{2.5}$-sec and ozone concentrations show single-peak variations, which appear higher in the daytime than at night with the peak occurring at about 15:00. As for each pollutant, the average maximum concentration appears higher for polluted stations than for clean ones, indicating that the atmospheric oxidation background is conducive to the formation of PM$_{2.5}$-sec.

**DATA AVAILABILITY STATEMENT**

The original contributions presented in the study are included in the article/Supplementary Material; further inquiries can be directed to the corresponding author.

**AUTHOR CONTRIBUTIONS**

XY contributed to data processing, mapping, analysis, and writing; YZ contributed to software and mapping; NL contributed to conceptualization and check on; and SY contributed to writing and editing.

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