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Spatio-temporal characteristics of PM_{2.5} and O₃ synergic pollutions and influence factors in the Yangtze River Delta

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Since the implementation of pollution prevention and control action in China in 2013, particulate pollution has been greatly reduced, while ozone pollution has become gradually severe, especially in the economically developed eastern region. Recently, a new situation of air pollution has emerged, namely, enhanced atmospheric oxidation, ascending regional ozone pollution, and increasing particle and ozone synergic pollution (i.e., double-high pollution). Based on the long-term observation data from 2015 to 2021, we examined the spatio-temporal characteristics of urban PM_{2.5} and O₃ pollution in the Yangtze River Delta and quantified the effects of meteorological and non-meteorological factors on pollution in four city clusters using stepwise multiple linear regression models. Temporally, PM_{2.5} decreased gradually year by year while, O₃ increased in city clusters. Spatially, PM_{2.5} declined from northwest to southeast, while O₃ decreased from northeast to southwest. Except for southern Zhejiang, other city clusters suffer from complex air pollution at different levels. In general, pollution intensity and frequency vary with city location and time. Single PM_{2.5} pollution mostly occurred in northern Anhui. Single O₃ pollution occurred in central and southern Jiangsu and northern Zhejiang. Synergic pollutions of PM_{2.5} and O₃ mainly occurred in central Jiangsu. The contributions (90%) of non-meteorological factors (e.g., anthropogenic emission) to PM_{2.5} decrease and O₃ increase are far larger than that of meteorological factors (5%). Relative humidity, sea level pressure, and planetary boundary layer height are the most important meteorological factors to drive PM_{2.5} changes during pollution. Downward solar radiation, total cloud cover, and precipitation are the most important meteorological factors that affect O₃ changes during pollution. The results provide insights into particulate and ozone pollution in the Yangtze River Delta and can help policymakers to formulate accurate air pollution prevention and control strategies at urban and city cluster scales in the future.

KEYWORDS

PM_{2.5}, ozone, pollution, meteorology, Yangtze River Delta

1 Introduction

The rapid economic growth in past decades has resulted in considerable industrialization, fast urbanization, and vast fossil fuel combustion in China, which emits large amounts of pollutants and then seriously harms air quality and the environment (Chan and Yao 2008; Chen et al., 2013; Sheehan et al., 2014; Qu et al., 2017; Li et al., 2018). The Chinese government has implemented several strict plans of clean air action and a series of control policies since 2012, such as the Action Plan for Prevention and Control of Air Pollution and the Winning Battle for Blue Sky (Chinese State Council 2013b; Chinese State Council, 2018), to reduce major pollutant concentrations, prevent air pollution, and improve air quality. Up to now, air quality has been drastically improved in economically developed areas in the eastern part of the country (Zheng et al., 2017; Zheng et al., 2018; Xue et al., 2019). However, the concentrations of particulate matter in some cities still fail to meet China's National Ambient Air Quality Standards (NAAQS) (MEE 2012) and are far from meeting the air quality guidelines recommended by the World Health Organization (WHO 2006). Meanwhile, ozone (O₃) shows an obvious upward trend and becomes the primary pollutant in most cities, including populous megacities (Li et al., 2017; Wang et al., 2019; Xu et al., 2019; Zeng et al., 2019). Numerous studies have proven that high levels of PM_{2.5} (particulate matter with aerodynamic diameters <2.5 μm) and O₃ are harmful to long-term exposed people, animals, and plants due to raising the risk of mortality (Pope III et al., 2006; Lepeule et al., 2012; Canella et al., 2016; Cohen et al., 2017; Poursafa et al., 2022). Additionally, environmental pollution is found to be a potential culprit for the high and progressively younger incidence of some human diseases (Xu et al., 2022). Recently, the synergic pollution of high concentrations of PM_{2.5} and O₃ has been frequently observed in China during agricultural biomass burning periods (Ding et al., 2013) and between late spring and early autumn (Tie et al., 2019), accompanying higher atmospheric oxidation and more secondary components (Zhang H. L. et al., 2015; Song et al., 2017). However, this new situation of air pollution is puzzling because the understanding of full chemical reactions is limited.

After years of pollution control efforts, the emission of primary particulate matter has dramatically reduced in China, and secondary generation dominates particulate matter origination in many areas (Wang et al., 2016). Although secondary PM_{2.5} and O₃ share common precursors, they have different formation mechanisms. O₃ is known to be a typical secondary component mainly formed by photochemical reactions of NO_x and volatile organic compounds (VOCs) in the presence of ultraviolet light (Pusede et al., 2015; Wang et al.,

2017). In fact, there exists a complex non-linear relationship between aerosol and O₃, that is, PM_{2.5} with sophisticated physicochemical properties affects the formation and loss of O₃ (Stadler et al., 2018; Li et al., 2019), and in turn, O₃ can impact atmospheric oxidation capacity and thus secondary aerosol formation (Pathak et al., 2009; Wang et al., 2016). Furthermore, air pollution is closely linked with climate change, and they interact through complex approaches in the atmosphere. For example, the changes in emissions and meteorological conditions have been confirmed to influence ambient pollutants together (He et al., 2003; Wang L. et al., 2015; Khuzestani et al., 2017). Using a multi-resolution emission inventory for China, Zheng et al. (2018) estimated nationwide changes of PM_{2.5}, SO₂, NO_x, and NMVOCs by -33%, -59%, -21%, and 2% between 2013 and 2017. Meteorological conditions can affect urban pollution (Tie et al., 2009), and their role varies with the terrain (Zhao et al., 2020). In the future, global climate change will exacerbate air pollution and cause unignorable climate-driven air pollution mortality (Hong et al., 2019).

O₃ as a major pollutant, has a significant impact on the air quality of the Yangtze River Delta (YRD), accounting for 55.4% of total days exceeding pollution standards in 2021 (accounting for 37.2% in 2015) (MEE 2015; MEE 2021). Daytime mean concentration of O₃ and its proportion acting as the major pollutant have increased significantly year by year due to rapid urban expansion in the YRD (Gu et al., 2011; Liao et al., 2015; Han et al., 2017). Previous studies focused on either particulate matter or O₃ pollution events at national or regional scales (Jiang et al., 2012; Tie et al., 2013; Gao et al., 2016; Ming et al., 2017; Shu et al., 2017; Dai et al., 2021), but paid less attention to their double-high pollution (DHP) episodes. Dai et al. (2021) investigated the DHP events in 25 cities of the YRD from 2013 to 2019 and found that they are mainly affected by high humidity, high surface temperature, and low wind speed. Qin et al. (2021) revealed the spatial distribution, trends, and meteorological characteristics of DHP periods in the YRD between 2015 and 2019. In this region, the pollutants show obvious spatial discrepancy due to different land use types, urban morphology, geographical location in city agglomerations (Zhang Q. et al., 2019; Mao et al., 2022), and strong seasonal variation and temporal correlation between cities within 250 km (Hu et al., 2014; Shen et al., 2017).

This paper used observation of the major pollutants in the YRD from 2015 through 2021 to explore the spatio-temporal pattern of PM_{2.5} and O₃ pollution. The goal is to characterize the pollution periods and their geographical differences from a regional city-cluster perspective and unravel the nature of PM_{2.5} and O₃ synergic pollutions and related drivers, including meteorological and other factors. The results will

provide insights into the current situation and upcoming challenges of air pollution and help policymakers to adopt further strategies for precise pollution control and prevention.

2 Materials and methods

2.1 Overview of city clusters

The YRD is one of the most economically developed and populous regions in China, with the largest urban agglomeration, including 41 cities, such as Shanghai (SH), Nanjing (NJ), Hangzhou (HZ), Suzhou (SZ), and Hefei (HF), etc. (Figure 1). The YRD has a complex land cover with plains mainly spreading in the north and east, and low and middle mountains in the southwest and south. We analyzed the correlation between $PM_{2.5}$ and O_3 in these 41 cities and then classified them as four city clusters, that is, Huaibei (HBC), Wanjiang (WJC), Jiangsu-northern Zhejiang-Shanghai (JZS), and southern Zhejiang (ZSC), based on high correlation coefficients ($R \geq 0.8$) and similarities in land cover (Figure 1). The spatial differences of $PM_{2.5}$ and O_3 concentrations are general small inside each city cluster but large between different city clusters. In addition, the city that holds the highest correlation coefficients with other cities in pollutant concentrations was selected as the proxy of one city cluster for the following analysis (Supplementary Figure S1). Four typical cities, Huaibei (HB), Hefei (HF), Wuxi (WX), and

Jinhua (JH), were selected as indicators for the above-mentioned city clusters (Table 1).

2.2 Data of air quality and meteorology

Hourly concentrations of O_3 , $PM_{2.5}$, CO, SO_2 , and NO_2 were taken from the archived data (<https://quotsoft.net/air>) of the Ministry of Ecology and Environment of China (Wang 2022). As part of the Clean Air Action Plan launched in 2013, the observation network on pollutants covers 496 sites in 74 major cities across the country (Chinese State Council 2013a) and has been extended to more than 2000 sites by 2021. The NAAQS guidelines are strictly abided by instrument operation and management, data assurance, and quality control, and SO_2 , NO_2 , and CO are monitored at the same sites as $PM_{2.5}$ (MEE 2012; Zhang and Cao 2015). Data used here span from January 2015 to December 2021, and their statistical validity is assessed by the Ambient Air Quality Standard (GB 3095-2012) and the trial Technical Specification for Ambient Air Quality Evaluation (HJ 663-2013) (Yang et al., 2020). The daily averages of $PM_{2.5}$, CO, SO_2 , and NO_2 were computed by their hourly averages at every site that contains more than 20 h of valid records in 1 day. The 8-h average of O_3 concentrations was calculated by its hourly values that possess at least six serial valid records within every 8 h. The maximum daily 8-h average (MDA8) O_3 concentration was determined by 8-h averages in 1 day that has more than 14 valid records from 8:00 to 24:00 local time (LT).

The meteorological data used in this study came from the fifth-generation European Center for Medium-Range Weather Forecasts atmospheric reanalysis (ERA5), with a horizontal resolution of $0.25^\circ \times 0.25^\circ$. The meteorological data in ERA5 are more accurate than other reanalysis data, notably for meteorological elements at the surface and in the low troposphere, and thus are currently employed in various scientific studies (Meng et al., 2018; Song et al., 2020). In order to minimize the error caused by interpolation, the neighboring method was used to match the *in-situ* observation data of air quality with the nearest grid data of ERA5 (Liu et al., 2015; Zhu and Yuan 2019). In light of the method of previous studies (Leung et al., 2018; Li et al., 2019; Chen et al., 2020), we used 26 meteorological parameters (Supplementary Table S1) as original candidate predictors for multiple linear regression (MLR) fitting, averaged over 24 h or daytimes (08:00–17:00 LT).

2.3 Estimation of secondary aerosols

The rapid increase of secondary particulate matter can lead to heavy pollution events with high $PM_{2.5}$ loadings (Dao et al., 2021). In addition, atmospheric precursors experience complex

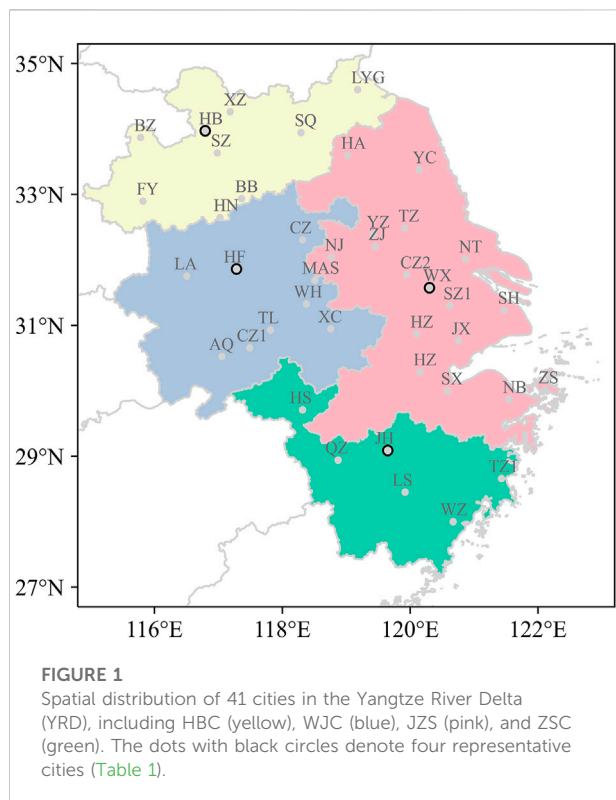


TABLE 1 Classification of urban agglomerations.

Clusters	Cities	Representative
Huaibei cluster (HBC)	Nine cities: Bozhou (BZ), Suzhou (SZ), Xuzhou (XZ), Huaibei (HB), Fuyang (FY), Huainan (HN), Bengbu (BB), Suqian (SQ), Lianyungang (LYG)	Huaibei (HB)
Wanjiang cluster (WJC)	Nine cities: Luan (LA), Anqing (AQ), Xuancheng (XC), Hefei (HF), Chuzhou (CZ), Chizhou (CZ1), Wuhu (WH), Tongling (TL), Maanshan (MAS)	Hefei (HF)
Jiangsu- northern Zhejiang-Shanghai cluster (JZS)	Seventeen cities: Huaian (HA), Nanjing (NJ), Suzhou (SZ1), Nantong (NT), Zhenjiang (ZJ), Jiaxing (JX), Ningbo (NB), Changzhou (CZ2), Yangzhou (YZ), Wuxi (WX), Hangzhou (HZ), Taizhou (TZ), Huzhou (HZ), Yancheng (YC), Shaoxing (SX), Shanghai (SH), Zhoushan (ZS)	Wuxi (WX)
Southern Zhejiang cluster (ZSC)	Six cities: Jinhua (JH), Huangshan (HS), Quzhou (QZ), Lishui (LS), Taizhou (TZ1), Wenzhou (WZ)	Jinhua (JH)

non-linear chemical reactions in secondary generation under favorable meteorological conditions. Analytical methods of real-time $PM_{2.5}$ monitoring cannot determine all secondary components. Chang and Lee (2007) used pollutant observations and employed CO as a tracer for primary emissions to estimate secondary $PM_{2.5}$ concentrations at different photochemical activities. The estimation of summertime secondary aerosols has been carried out in many cities in China (Cui et al., 2013; Jia et al., 2017; Li H. L. et al., 2020; Gu et al., 2022; Yao et al., 2022; Yu et al., 2022), and research has proven that the method of CO tracer is able to screen primary $PM_{2.5}$ and estimate urban-scale secondary $PM_{2.5}$ concentration. The formation of secondary particulate matter links closely with photochemical activity. In particular, O_3 has commonly acted as an index of photochemical reactions to quantify the role of secondary particulate matter in air quality changes (Turpin and Huntzicker 1995; Na et al., 2004; Chang and Lee 2007; Wang Z. S. et al., 2015). Taking CO as a primary tracer and assuming that the structure of the emission source remains essentially stable, the larger the $PM_{2.5}/CO$, the larger the proportion of secondary- $PM_{2.5}$ (Chang and Lee 2007; Zhang Q. Y. et al., 2015). We divided the photochemical activity into four groups based on the daily hourly maximum of O_3 concentration ($O_{3,max}$): low for below $100 \mu g m^{-3}$, moderate for $100-160 \mu g m^{-3}$, high for $160-200 \mu g m^{-3}$, and very-high for above $200 \mu g m^{-3}$. The primary aerosols were estimated using hourly CO concentrations under different photochemical activity levels. Meanwhile, the secondary aerosols were calculated by deducting primary $PM_{2.5}$ from observed $PM_{2.5}$ (Cui et al., 2013; Li K. et al., 2020; Yu et al., 2022).

$$(PM_{2.5})_{s,i} = (PM_{2.5})_{t,i} - CO_{t,i} \times \left(\frac{PM_{2.5}}{CO} \right)_{low} \quad (1)$$

where, i ($i = 1,2,3$) denotes moderate, high, and very-high photochemical activity levels, respectively. $(PM_{2.5})_s$ represents secondary $PM_{2.5}$ concentration. $(PM_{2.5})_t$, and CO_t represent total

concentrations of $PM_{2.5}$ and CO in the atmosphere. $(PM_{2.5}/CO)_{low}$ refers to the 25th percentile of hourly ratios for $PM_{2.5}/CO$ at a low photochemical activity.

2.4 Stepwise multiple linear regression model

To quantify the influence of meteorology on air quality, we developed a stepwise MLR model to establish the relationship between pollutant concentrations and meteorological variables. The MLR model has been successfully applied in evaluating meteorological effects on $PM_{2.5}$ and O_3 changes (Tu et al., 2007; Tai et al., 2010; Xu et al., 2011; Yang et al., 2016; Wang et al., 2017; Zhao and Wang 2017; Yang et al., 2019; Zhai et al., 2019).

$$C_{i,p,c}(t) = b_{0,i,p,c} + \sum_{k=1}^N b_{k,i,p,c} \times Met_k(t) + \epsilon \quad (2)$$

where $C_{i,p,c}(t)$ is the observed daily pollutant i concentration ($PM_{2.5}$ or MDA8 O_3) at period p and city c , $Met_k(t)$ is one of the N meteorological predictors, b_0 is the intercept term, b_k is the regression coefficient of the k -th meteorological predictor, and ϵ is the residual term.

This study used the method of Chen et al. (2020) to obtain the best meteorological predictor. We minimized the effect of correlations between predictor variables using variance inflation factor (VIF) (Altland 1999; Che et al., 2019; Li H. L. et al., 2020) and based on the Akaike Information Criterion (AIC), adding or deleting statistical predictor variables to obtain the best fitting model when AIC reaches a minimum (Kutner et al., 2004). Supplementary Tables S2, S3 present optimal meteorological variables, calculated intercepts (b_0), regression coefficients (b_k), and adjusted coefficients of determination (R^2) for each city and period. The calculated adjusted R^2 , which reflects the fraction of variability described by MLR, is 0.2–0.6 for $PM_{2.5}$ and 0.5–0.8 for MDA8 O_3 , indicating the MLR model works reasonably well.

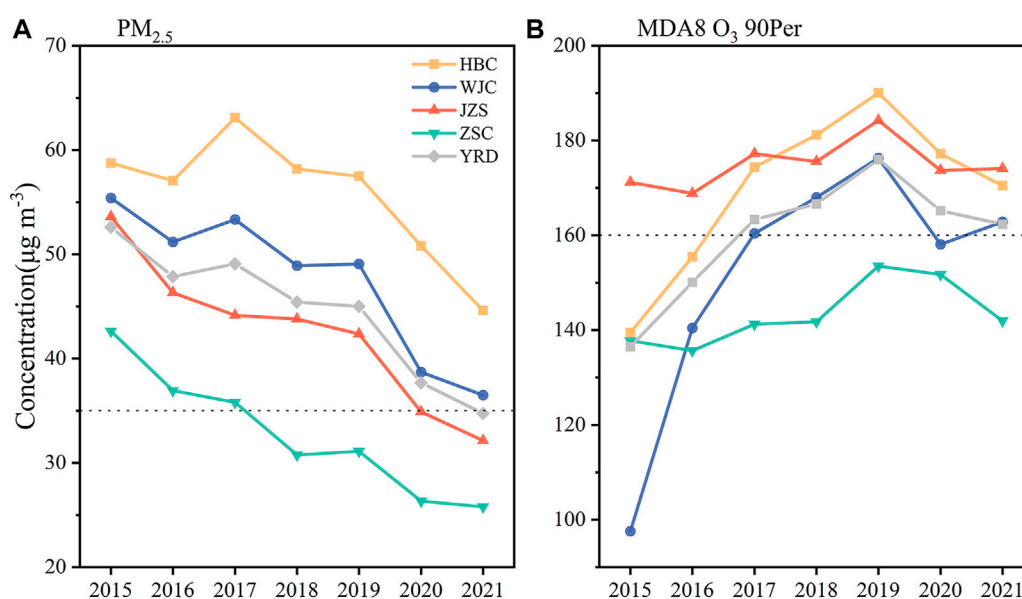


FIGURE 2

Inter-annual variations of (A) mean $PM_{2.5}$ and (B) MDA8 O_3 90Per in four city clusters and the YRD during 2015–2021. Black dotted lines denote the Class II of air quality standards (GB3095-2012).

2.5 Estimation of meteorological and non-meteorological contributions

The meteorological-driven changes (or trends) of pollutant concentrations (ΔP) were calculated directly from the predicted concentration ($P(t)$) after the MLR model was established:

$$\Delta P_{i,s,c} = \sum_{k=1}^N b_{k,i,s,c} \times \Delta \text{Met}_k \quad (3)$$

where ΔMet_k denotes the change (or trend) of the k -th meteorological variable. Non-meteorological driven changes (or trends) are mainly attributed to changes in anthropogenic emissions (Akaike 1969; Seo et al., 2018) and can be obtained from the difference between observed (ΔC) and meteorological-driven (ΔP) values. The relative contribution of each meteorological variable to total meteorological-driven changes (or trends) was quantified by the ratio of ($b_k \times \Delta \text{Met}_k$) to ΔP . The meteorological variable with the largest contribution was considered the meteorological variable with the largest impact on $PM_{2.5}$ and O_3 changes or trends.

3 Results and discussion

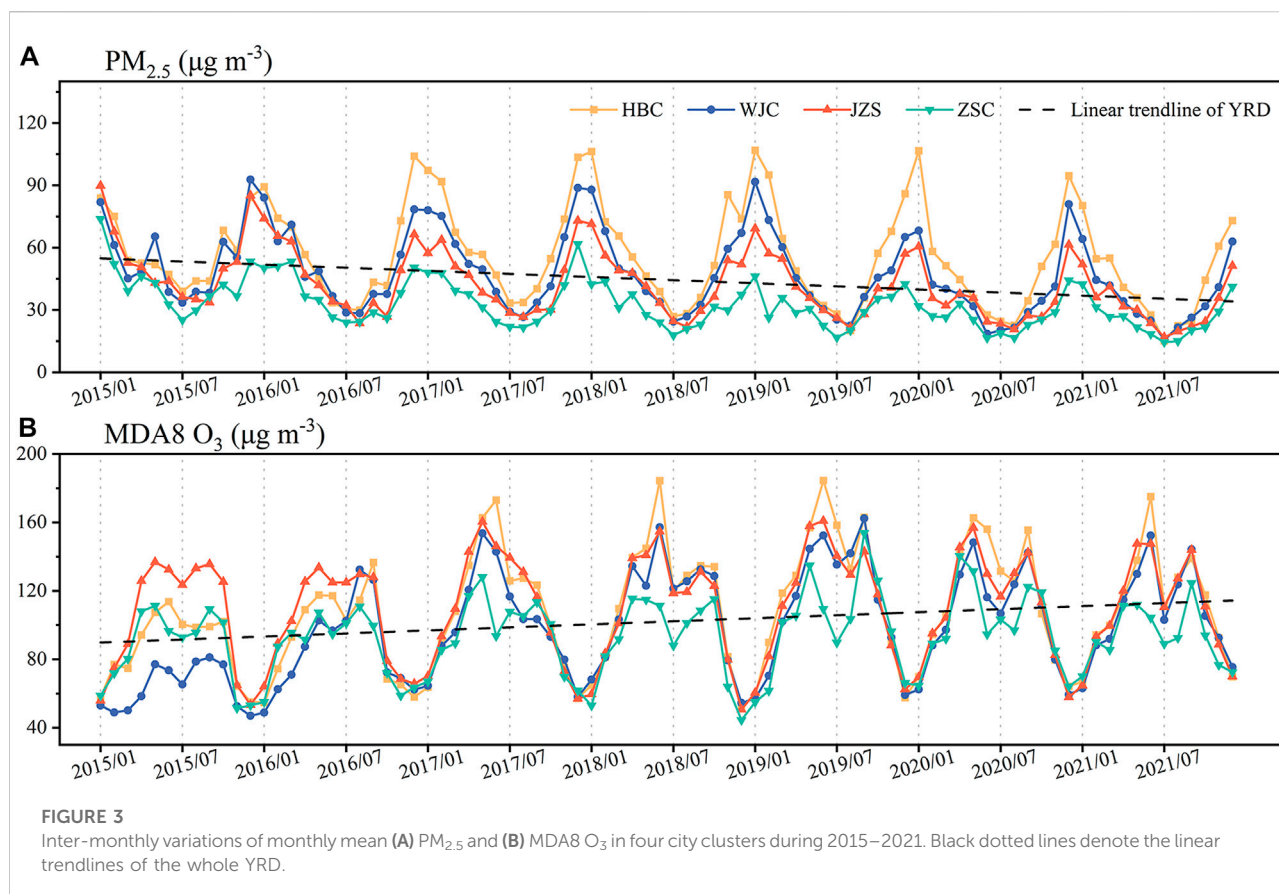
3.1 Characteristics of $PM_{2.5}$ and O_3 pollutions

3.1.1 Trends of $PM_{2.5}$ and O_3 concentrations

In almost all cities of the YRD, $PM_{2.5}$ shows a downward trend from 2015 through 2021 (Supplementary Figure S2), and

the proportion of cities with $PM_{2.5}$ exceeding the standard (ES) of air quality (annual mean $PM_{2.5}$ larger than $35 \mu\text{g m}^{-3}$) descends from 95.1% to 51.2%. The annual mean of $PM_{2.5}$ concentration was highest ($66 \mu\text{g m}^{-3}$) in Hefei in 2015 and in Fuyang ($49 \mu\text{g m}^{-3}$) in 2021. Meanwhile, MDA8 O_3 shows an upward trend in 2015–2019 and then a downward trend in 2020–2021, and the proportion of cities with MDA8 O_3 ES of air quality (MDA8 O_3 90Per larger than $160 \mu\text{g m}^{-3}$) in total cities ascends from 41.5% to 80.5% and then descends to 68.3%. The 90Per of MDA8 O_3 was the highest ($196 \mu\text{g m}^{-3}$) in Yangzhou in 2015 and in Changzhou ($195 \mu\text{g m}^{-3}$) in 2021. On the other hand, in terms of spatial distribution pattern, $PM_{2.5}$ generally reduced from northwest to southeast, while O_3 reduced from northeast to southwest.

Figure 2 depicts the inter-annual variations of $PM_{2.5}$ and O_3 concentrations in the YRD. $PM_{2.5}$ decreased by 24.1%, 34.1%, 40.1%, and 39.1% between 2015 and 2021 in HBC, WJC, JZS, and ZSC city clusters, respectively, especially in the JZS, with the most notable reduction. Obviously, $PM_{2.5}$ in the HBC city cluster is much higher than others (Figure 2A), and most cities in this cluster suffer from severe particle pollution because of large coal consumption as they are located in coal-producing areas. In contrast, $PM_{2.5}$ in the ZSC city cluster is far lower than that in other city clusters due to natural vegetation coverage (Figure 2A). In particular, Lishui, Wenzhou, and Taizhou have already met the NAAQS. $PM_{2.5}$ of the HBC city cluster increased slightly before 2017 (Figure 2A). In fact, most cities in Anhui province showed an increase of $PM_{2.5}$ at the same time, especially in Fuyang, Huaibei, and Suzhou. It is worth noting that Xuzhou,



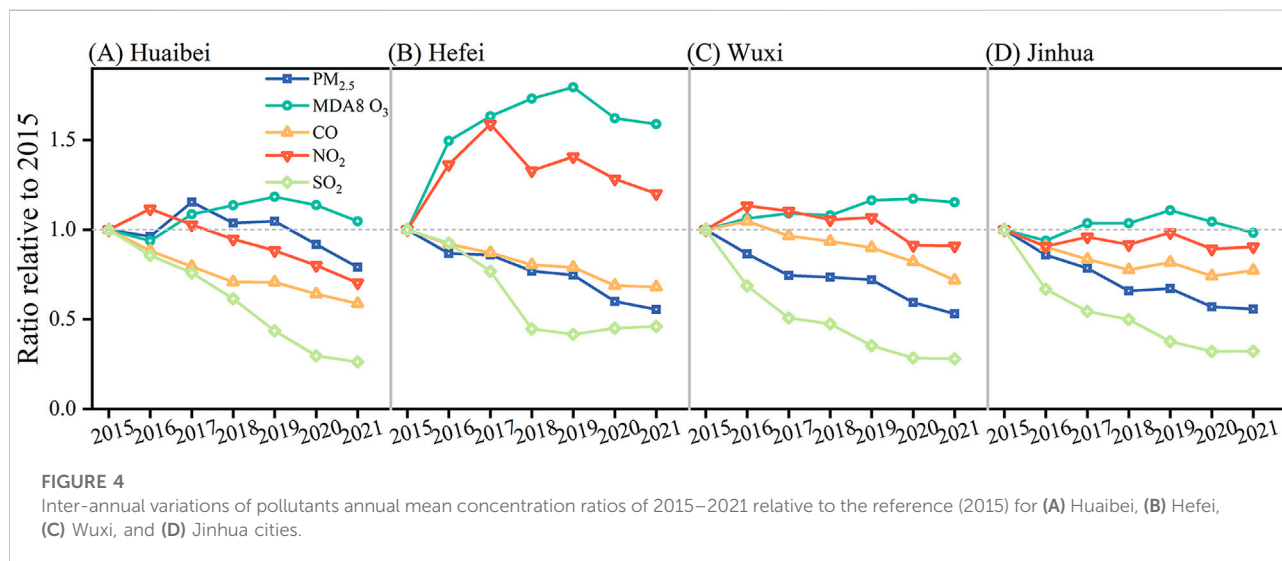
situated at the junction of Jiangsu, Shandong, Henan, and Anhui provinces, is vulnerable to non-local pollutants transported from remote sources and thus has a relatively higher $\text{PM}_{2.5}$ level than surrounding cities. On the other hand, Xuzhou is the only southern city that is allowed to provide centralized heating in winter, which is bound to coal-fired emission pollution (Wang et al., 2020).

MDA8 O_3 shows a tipping point in 2019, with a rising trend before 2019 and a falling trend afterward (Figure 2B). The increasing rates were 22.2%, 66.9%, 1.7%, and 3% in the HBC, WJC, JZS, and ZSC city clusters, respectively, from 2015 through 2021. The MDA8 O_3 in the JZS city cluster exceeded the NAAQS and kept fluctuating at a high level (Figure 2B), indicating that the urban group has been suffering from serious O_3 pollution for a long time. By contrast, the rising trend of MDA8 O_3 in the ZSC city cluster was insignificant, and all cities met the NAAQS (Figure 2B). The most notable increase of MDA8 O_3 occurred in the WJC city cluster, especially in 2016, with an increase of $43 \mu\text{g m}^{-3}$ (Figure 2B), which is mainly attributed to anthropogenic emissions. On the whole, the ZSC city cluster has the best air quality in the YRD, whereas others have varying severity of combined $\text{PM}_{2.5}$ and O_3 pollution.

Figure 3 presents inter-monthly variations of mean $\text{PM}_{2.5}$ and MDA8 O_3 concentrations. $\text{PM}_{2.5}$ and MDA8 O_3 show

contrary long-term trends on a monthly-scale. Previous studies have found that high particulate matter loadings usually match low O_3 concentrations in the cold season (Sun L. et al., 2019), and low particulate matter loadings match high O_3 concentrations in the warm season (Li et al., 2016). $\text{PM}_{2.5}$ generally exhibits high levels in winter and low levels in summer due to fossil fuel combustions in the cold season and turbulent vertical mixing (Ding et al., 2013; Yue et al., 2015; Zhang et al., 2018), usually reaching peaks in December or January and troughs in July, with a single-peak-valley pattern. The increased emissions during the winter heating period in North China can exacerbate particulate matter pollution in the Yangtze River Delta through the long-range transport of pollutants (Zhang H. L. et al., 2015; Zhao et al., 2015). The monthly $\text{PM}_{2.5}$ of the ZSC city cluster fluctuates slightly around $40 \mu\text{g m}^{-3}$, far less in magnitude compared with other city clusters (Figure 3A). Meanwhile, $\text{PM}_{2.5}$ in these city clusters had almost no difference in the first 2 years (2015–2016), but an obvious difference in the following 5 years (2017–2021), especially in the months before and after the peak. Except for July and August, the median $\text{PM}_{2.5}$ of all city clusters is higher than $35 \mu\text{g m}^{-3}$, indicating that most cities in the YRD suffer from $\text{PM}_{2.5}$ pollution to some extent.

MDA8 O_3 continues to rise with double-peak-valley fluctuations, with the peaks in May or June and September,



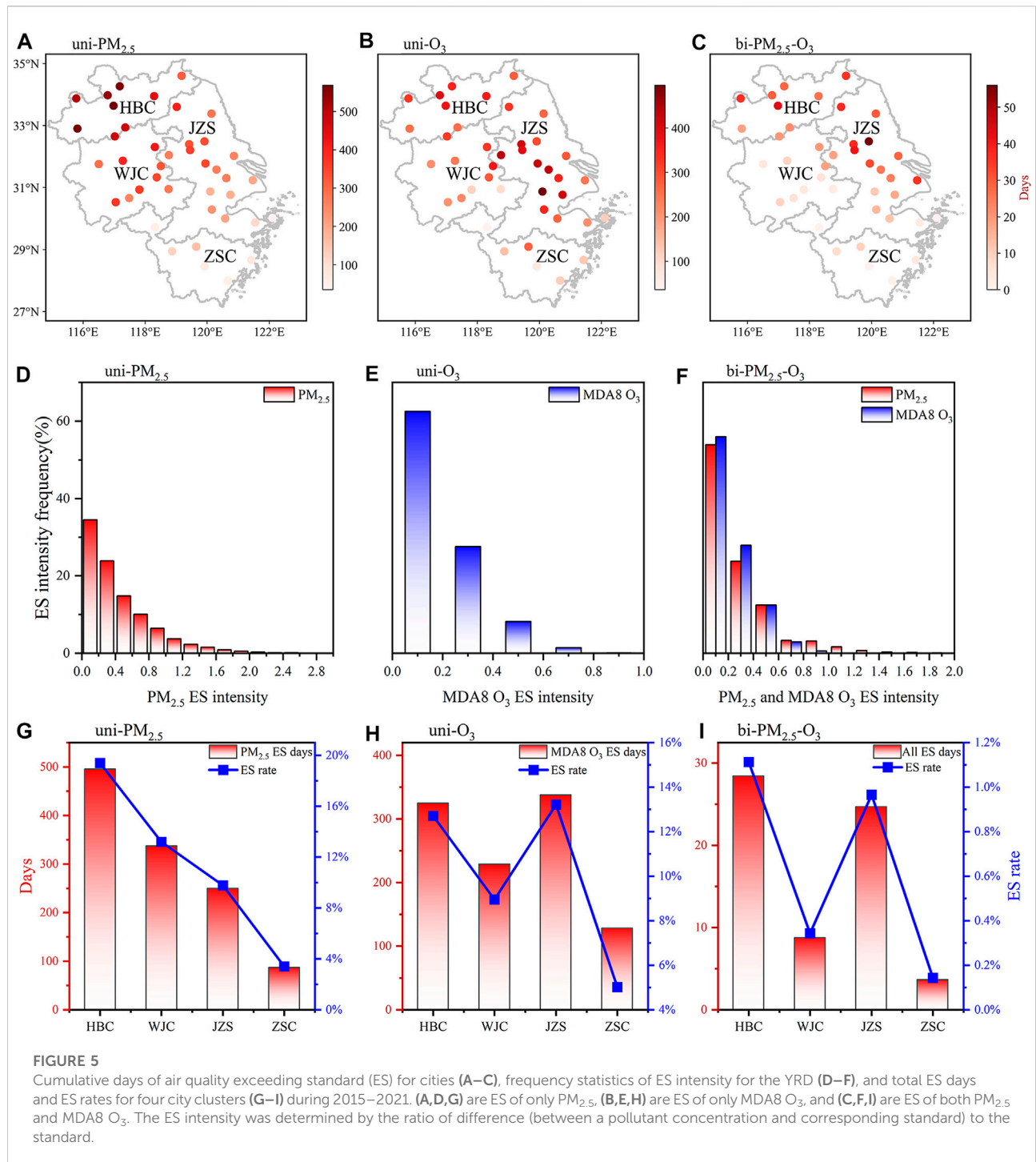
and the troughs in July and December (Figure 3B). The closer to the peak month, the greater the difference in concentrations among four city clusters. High temperature, low relative humidity and intense solar radiation enhance O₃ formation in summer, whereas the meteorological conditions unfavorable to photochemistry and increasing NO titration suppress O₃ yield (Yang et al., 2021).

Figure 4 shows the ratios of major pollutants from 2015 through 2021 relative to 2015 as the reference in Huaibei, Hefei, Wuxi, and Jinhua cities. PM_{2.5} exhibited a consistent decline in Hefei, Wuxi, and Jinhua. However, it rose dramatically before 2017 and then turned to a continuous decline in Huaibei (Figure 4A). In all four cities, MDA8 O₃ showed an increase before 2019 and subsequently decreased, a time point of COVID-19 occurrence (the beginning of December 2019). Hefei has the greatest increase from 2015 through 2019, with an increment of almost 80% (Figure 4B), far higher than the growth of the other three cities. NO₂ showed long-term changes similar to O₃. On the whole, SO₂ reduced year by year at an average range of 53.9%–73.7%, and CO gradually decreased by 22.6%–41.2%. The reasonable explanation is the emission reduction of primary pollutants and related precursors by the implementation of the air cleaning plan since 2013, including efficient control methods such as tightening industrial emissions, modernizing industrial boilers, retiring outdated industrial capacity, and encouraging clean fuels in the residential sector (Lang et al., 2017; Shao et al., 2018; Zhai et al., 2019; Zhang T. et al., 2019). Since the outbreak of COVID-19, human activities were reduced due to disease control regulations that lasted throughout 2020 and 2021 (Wang and Zhang 2020).

3.1.2 Intensity of PM_{2.5} and O₃ pollutions

According to the Class II of NAAQS, we defined the single particulate matter pollution (uni-PM_{2.5}) as the day with only

daily PM_{2.5} exceeding 75 μg m⁻³, the single O₃ pollution (uni-O₃) as the day with only MDA8 O₃ exceeding 160 μg m⁻³, and the synergic pollution of particulate matter and O₃ (bi-PM_{2.5}-O₃) as both PM_{2.5} and MDA8 O₃ above the aforementioned standards in a day (i.e., DHP pollution). Figures 5A–C shows the cumulative days of uni-PM_{2.5}, uni-O₃ and bi-PM_{2.5}-O₃ in the YRD from 2015 through 2021. Overall, from a spatial perspective, the total number of uni-PM_{2.5} days decreased gradually from northwest to southeast in the YRD region, with the maximum number of days found in the north, including northern Anhui and northern Jiangsu provinces (Figure 5A). The uni-O₃ increased from northeast to southwest, with the maximum number of days in the central YRD, including southern Jiangsu, Shanghai, and northern Zhejiang (Figure 5B). The bi-PM_{2.5}-O₃ days mainly occurred in northern Anhui and Jiangsu provinces, especially on the line from Bozhou to Shanghai (Figure 5C). From seasonal perspective, the uni-PM_{2.5} days mostly occurred in winter (November–February). However, the uni-O₃ days mainly appear in warm seasons (April–September), with two peaks in May, June and September (Supplementary Figure S3). The bi-PM_{2.5}-O₃ days mainly occur from late spring to early summer and late autumn, similar to O₃ pollution, with more distinct and obvious two peaks in April and October (Supplementary Figures S3, S4). Serious bi-PM_{2.5}-O₃ pollution always occurred in the cities in central and southern Jiangsu province in April, whereas in the cities in northern Anhui province, bi-PM_{2.5}-O₃ pollution occurred in October (Supplementary Figure S4). The uni-PM_{2.5} days significantly decreased year by year in every city, especially in the HBC and WJC city clusters (Supplementary Figure S5A). Except for the ZSC city cluster, the uni-O₃ days increased in the other three city clusters, peaked in 2019, and then decreased until 2021 (Supplementary Figure S5B). Among them, the WJC city



cluster has the most rapid growth, indicating an increasing severity of O₃ pollution in recent years. The bi-PM_{2.5}-O₃ days have prominent inter-annual changes and appear notably in 2015 and 2018 (Supplementary Figure S5C).

Figures 5D–F show the frequency statistics of pollution intensity of uni-PM_{2.5}, uni-O₃, and bi-PM_{2.5}-O₃ in YRD from 2015 through 2021. PM_{2.5} concentrations were higher

during uni-PM_{2.5} than bi-PM_{2.5}-O₃. However, opposite to particulate matter, MDA8 O₃ concentrations were lower during uni-O₃ than bi-PM_{2.5}-O₃, which is similar to the results of Qin et al. (2021) and Awang et al. (2018). Figures 5G–I present the cumulative polluted days and the percentages of total ES days in city clusters from 2015 through 2021. The HBC city cluster has the most

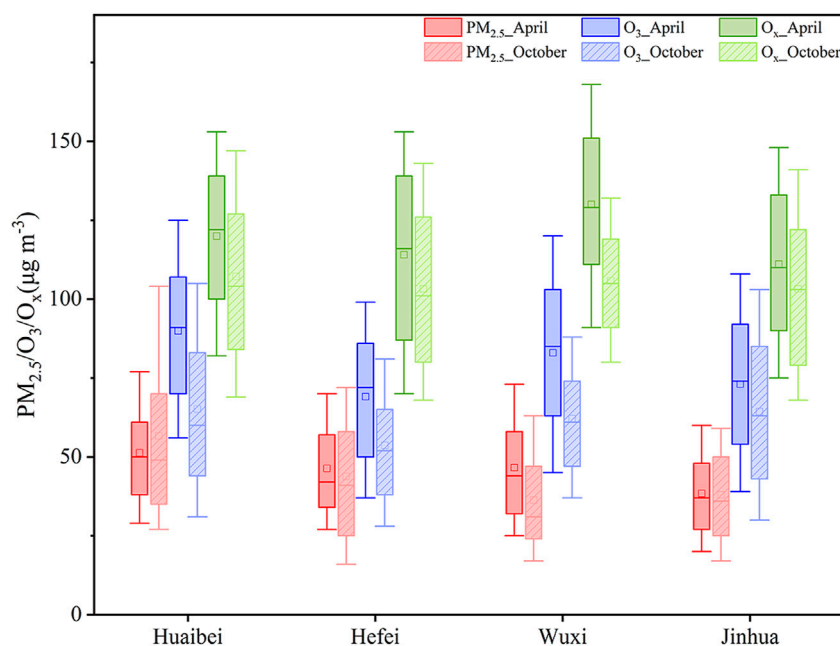


FIGURE 6

$PM_{2.5}$, O_3 , and $O_x (=O_3+NO_2)$ concentrations of Huaibei, Hefei, Wuxi, and Jinhua cities in April and October from 2015 through 2021. The upper and lower boundaries of the box represent the 75th and 25th percentiles, respectively. The short line within the box represents the median. The whiskers represent the 10th and 90th percentiles. The square represents the average.

serious uni- $PM_{2.5}$, with a total of 496 polluted days (19.4%). The JZS and HBC city clusters exhibit the most severe uni- O_3 , with percentage of 13.2% (338 days) and 13.7% (325 days). Meanwhile, the HBC and JZC city clusters have the worst bi- $PM_{2.5}$ - O_3 pollution, with a total of 28 days (1.1%) and 25 days (1%). Qin et al. (2021) reported that the highest frequency of bi- $PM_{2.5}$ - O_3 pollution appeared in Shanghai and the lowest in Anhui in the YRD. This inconsistency with our results may be related to the discrepancy in the pollution region divisions concerned.

3.2 Potential influence between $PM_{2.5}$ and O_3

Atmospheric oxidation refers to the ability of the atmosphere to remove pollutants through oxidation reactions, and strong atmospheric oxidation can promote the formation of secondary pollutants and particle aging (Levy 1971). In this study, we introduced O_x ($O_x = O_3 + NO_2$) to describe the atmospheric oxidative capacity (Cheung and Wang 2001; Clapp and Jenkin 2001; Herndon et al., 2008; Zhang et al., 2012), and to analyze the relationship between O_3 and atmospheric oxidation. $PM_{2.5}$ concentrations were almost the same in four typical cities in April and October, but O_3 , O_3/O_x ratio, and the Pearson correlation coefficients between O_3 and O_x were generally

higher in April than those in October (Figure 6; Table 2), indicating that O_3 had a more obvious contribution to atmospheric oxidation in April and varied significantly between cities. The O_3/O_x ratios greater than 0.5 indicated that atmospheric oxidation was mainly dominated by O_3 .

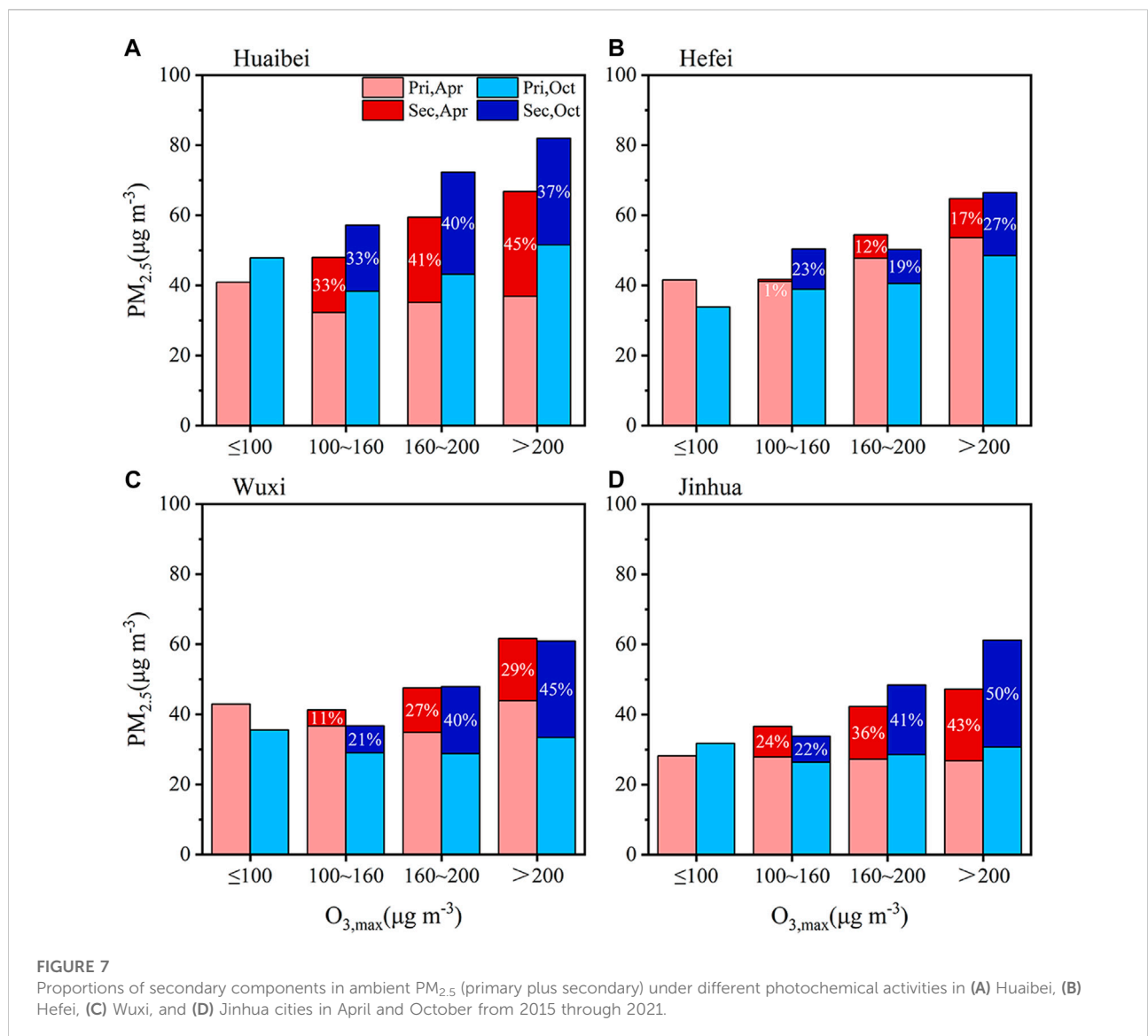
Figure 7 shows the proportions of estimated secondary components in $PM_{2.5}$ for four typical cities in April and October under different photochemical activity levels. The insignificant changes in primary $PM_{2.5}$ indicated that the structure of primary emission sources in these cities remained stable. The proportions of secondary particulate matter in October were generally greater than that in April in Hefei, Wuxi, and Jinhua cities, but it was the opposite in Huaibei city, with high photochemical activities. Hefei city had the smallest proportion of secondary particulate matter. The higher the photochemical activity levels, the larger the proportion of secondary components in $PM_{2.5}$. The results indicate that the generation and accumulation of secondary particulate matter have increasing significance on $PM_{2.5}$ concentrations and demonstrate that secondary $PM_{2.5}$ links closely with O_3 in a synergistic manner.

Based on the observation data of cities in the YRD, $PM_{2.5}$ was divided into low ($\leq 35 \mu g m^{-3}$), medium ($35-75 \mu g m^{-3}$), and high ($> 75 \mu g m^{-3}$) levels. Figure 8 shows the diurnal variations of the O_3 change rate calculated using hourly O_3 concentrations at the above three $PM_{2.5}$ levels. The daily trends of O_3 change rates were

TABLE 2 O₃, O_x (O_x = O₃ + NO₂) averages and ratios of O₃/O_x as well as correlations (R) between O_x and O₃ in April and October over 2015–2021 in Huaibei, Hefei, Wuxi, and Jinhua.

Huaibei					Hefei			
Month	O ₃	O _x	O ₃ /O _x	R	O ₃	O _x	O ₃ /O _x	R
April	89.9	120	0.75	0.95**	69.1	114	0.61	0.87**
October	65.2	107.2	0.61	0.91**	53.6	103.2	0.52	0.79**
Wuxi					Jinhua			
Month	O ₃	O _x	O ₃ /O _x	R	O ₃	O _x	O ₃ /O _x	R
April	83	130	0.64	0.89**	73	111.1	0.66	0.93**
October	62.1	105.8	0.59	0.77**	64.3	103.2	0.62	0.91**

**Passing the significant levels $p < 0.01$.



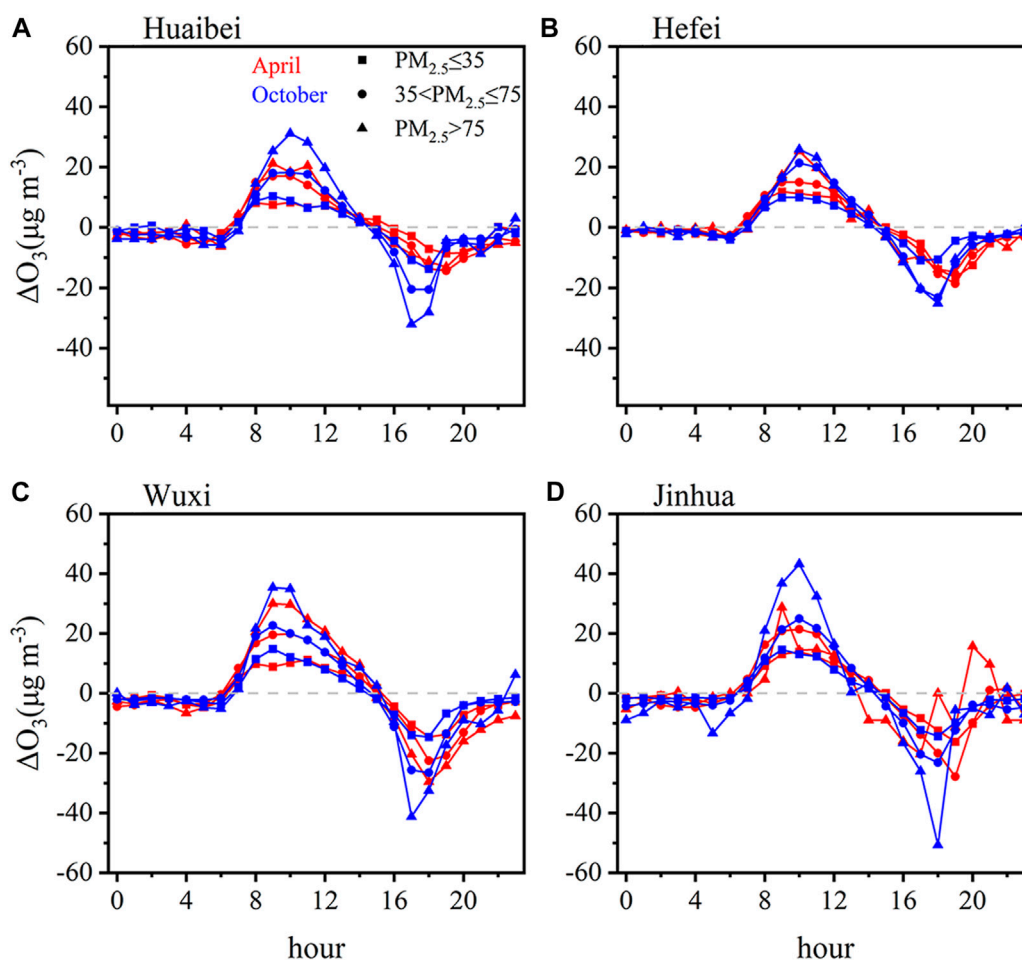


FIGURE 8

Diurnal variations of O_3 increment (next minus previous) at three $PM_{2.5}$ levels in (A) Huaibei, (B) Hefei, (C) Wuxi, and (D) Jinhua cities in April and October from 2015 through 2021. Positive and negative indicate the production and loss of ozone.

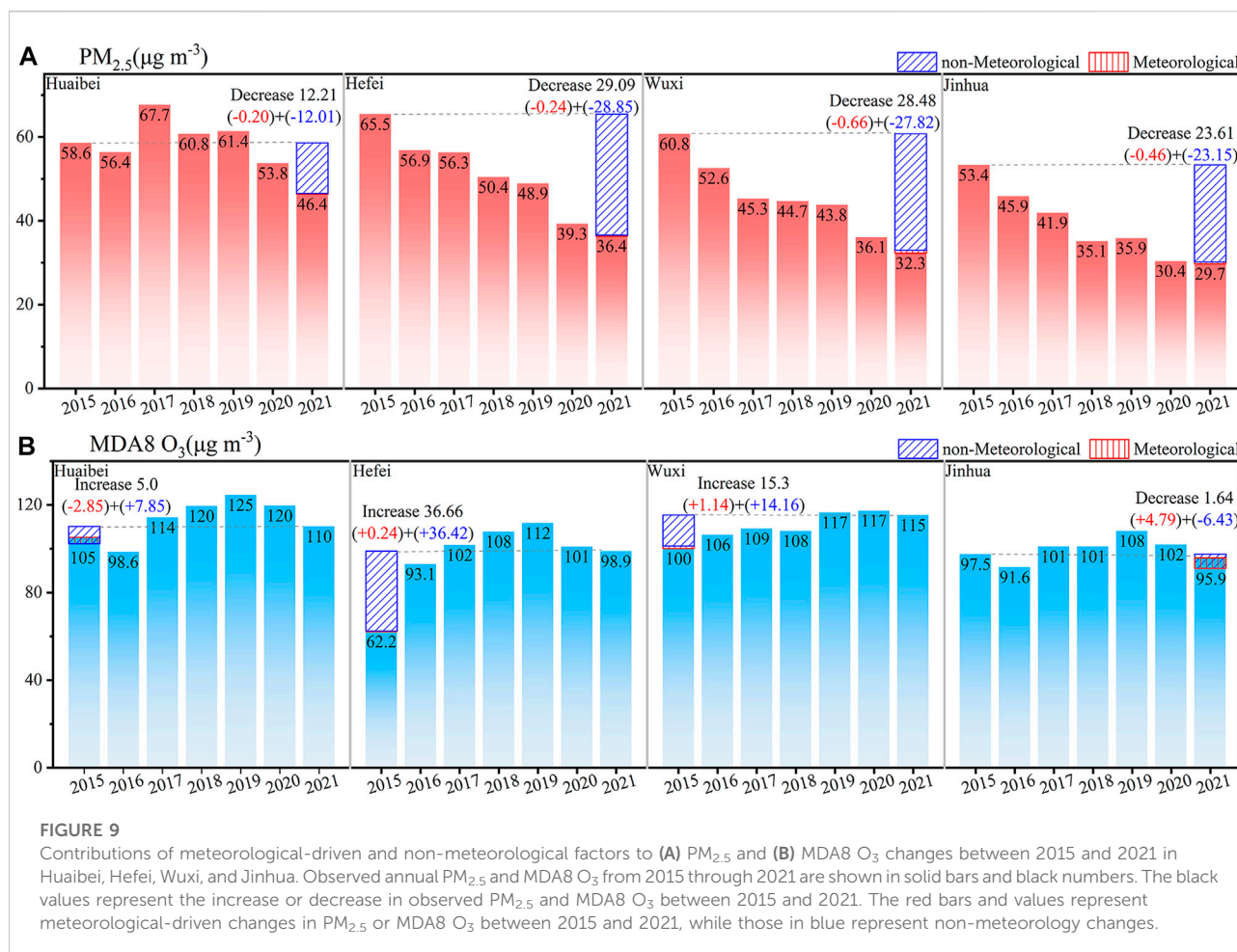
almost the same in the four cities at different $PM_{2.5}$ levels, that is, zero from 22:00 to 7:00 LT, positive from 7:00 to 16:00 LT, and negative from 16:00 to 22:00 LT. The daily variations of O_3 concentrations exhibited a single-peak-valley pattern, with the peak occurring at 11:00–12:00 LT and the trough at 17:00–18:00 LT. This was probably due to changes in solar radiation, traffic emissions, the amount of NO_x and VOC_s precursors, NO titration effect on O_3 consumption, photochemical reactions, etc. Several studies (Deng et al., 2011; Cai et al., 2013; Zhao et al., 2018) have shown that high concentrations of $PM_{2.5}$ absorb and weaken total solar radiation, reducing the rate of photochemical reactions and thus inhibiting O_3 production. While Figure 8 depicts that with the increase of $PM_{2.5}$ loadings, the peak range of O_3 variability gradually broadens, implying that the increase of $PM_{2.5}$ concentration in these cities can promote O_3 production to some extent in April and October. Additionally, Chi (2018) and Zhu (2018) discovered that an increase in aerosol raises the

concentration of O_3 in conditions of clear skies and light pollution. This demonstrates once more that there is no straightforward linear link between aerosols and O_3 concentration.

3.3 Driver contributions to $PM_{2.5}$ and O_3 pollutions

3.3.1 Meteorological and non-meteorological contributions

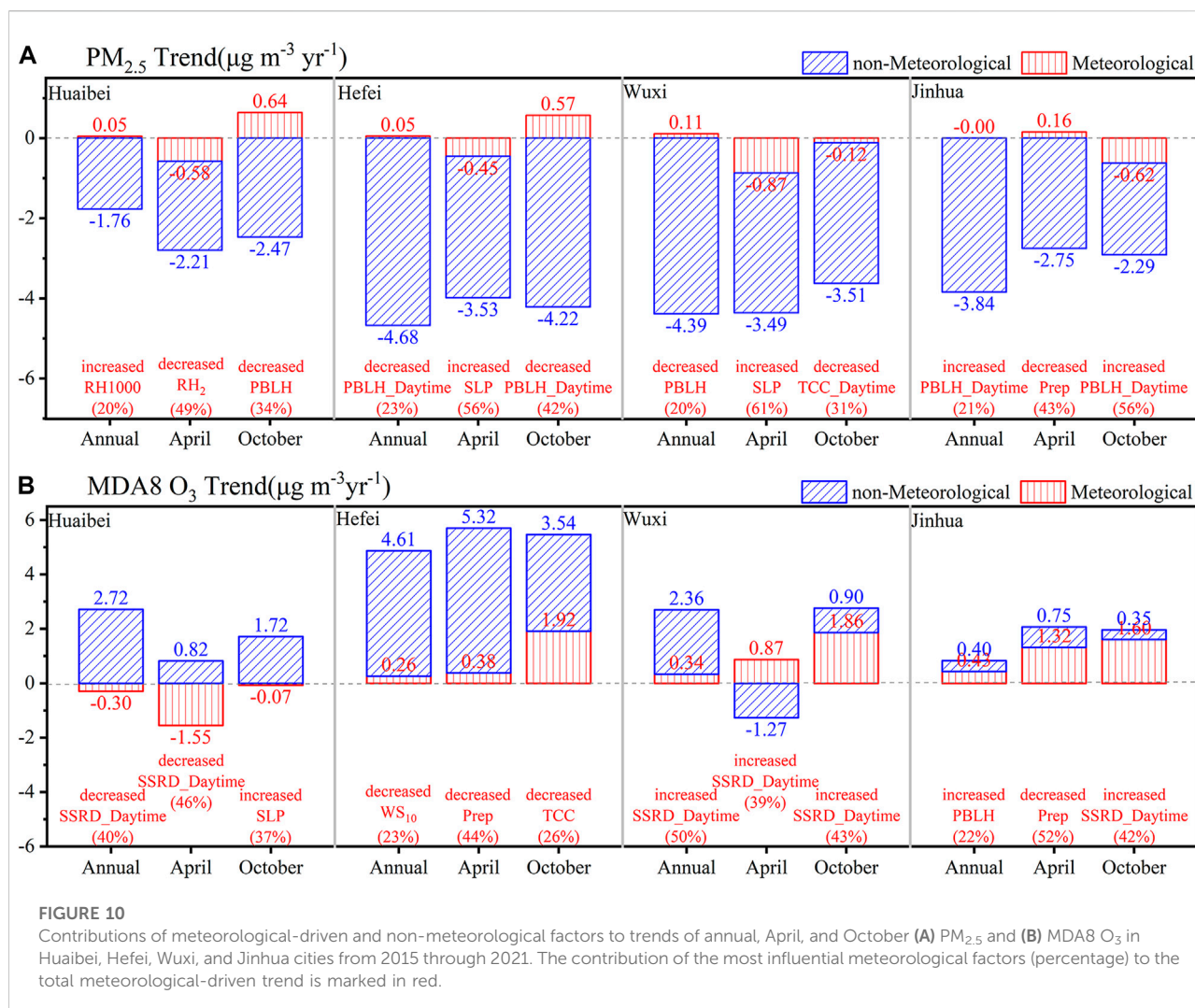
Figure 9A shows the changes in annual mean $PM_{2.5}$ concentrations from 2015 through 2021 in four typical cities. A significant decline of annual mean $PM_{2.5}$ was observed in Huaibei, Hefei, Wuxi, and Jinhua cities, with a reduction of $12.21 \mu g m^{-3}$, $29.09 \mu g m^{-3}$, $28.48 \mu g m^{-3}$, and $23.61 \mu g m^{-3}$ between 2015 and 2021, respectively. Among them, the



contributions of non-meteorological changes to $PM_{2.5}$ decrease were estimated to be $12.01 \mu g m^{-3}$, $28.85 \mu g m^{-3}$, $27.82 \mu g m^{-3}$, and $23.15 \mu g m^{-3}$, respectively, accounting for more than 90% of the total decrease. Zhang X. et al., 2019 reported that the improvement of air quality on $PM_{2.5}$ is primarily due to anthropogenic emission reductions of SO_2 , NO_x , BC, OC, and primary particles. Therefore, implementing pollution control measures is crucial to lowering $PM_{2.5}$ loadings and reducing pollution (Zhang Q. et al., 2019; Chen et al., 2020; Li K. et al., 2020). The $PM_{2.5}$ of Huaibei decreased the most because of a significant increase in 2017 due to massive anthropogenic emissions (Supplementary Figure S6A). Compared with 2015, the $PM_{2.5}$ of Huaibei, Hefei, Wuxi, and Jinhua cities decreased by $0.2 \mu g m^{-3}$, $0.24 \mu g m^{-3}$, $0.66 \mu g m^{-3}$, and $0.46 \mu g m^{-3}$ due to changes in meteorological conditions, accounting for less than 5% of the total reduction (Figure 9A). Weather and climatic changes are conducive to $PM_{2.5}$ reduction, but do not lead to a substantial improvement in air quality (Zhang T. et al., 2019). However, in specific years, for example, Huaibei in 2016, Wuxi in 2018, and Hefei in 2019, the impact of meteorological conditions on $PM_{2.5}$ outweighed that of anthropogenic emissions

(Supplementary Figure S6A). Therefore, the inter-annual variability in meteorology must be considered in designing future control strategies to improve air quality (Ding et al., 2019).

Figure 9B shows the annual mean $MDA8 O_3$ from 2015 through 2021 for four typical cities. The average $MDA8 O_3$ increased significantly by $5 \mu g m^{-3}$, $36.66 \mu g m^{-3}$, and $15.3 \mu g m^{-3}$ in Huaibei, Hefei, and Wuxi cities, respectively, but it decreased by $1.64 \mu g m^{-3}$ in Jinhua. The non-meteorological change contributed to the increase of $MDA8 O_3$ by $7.85 \mu g m^{-3}$, $36.42 \mu g m^{-3}$, and $14.16 \mu g m^{-3}$ in Huaibei, Hefei, and Wuxi cities, accounting for 73%, 99%, and 93% of total changes, respectively. $MDA8 O_3$ in Jinhua decreases by $6.43 \mu g m^{-3}$ due to non-meteorological changes (Figure 9B). O_3 pollution was possibly dominated by the NO_x -limited mechanism due to good vegetation coverage in local and surrounding regions. Previous studies have suggested that one reason for the O_3 increase is that low $PM_{2.5}$ loadings reduce sunlight scattering and absorption in the atmosphere, increasing ultraviolet radiation arriving at the ground and leading to high O_3 concentrations (Dickerson et al., 1997; Li et al., 2011; Tao et al., 2014). In addition, slowing aerosol sinks of hydrogen



peroxide radicals can promote O₃ formation (Li et al., 2019). Furthermore, increasing VOCs emissions and reducing NO_x titration over urban areas can result in high O₃ concentrations (Fu et al., 2019; Sun W. W. et al., 2019). As shown in Figure 9B, the meteorological-driven changes in MDA8 O₃ are estimated to be $-2.85 \mu\text{g m}^{-3}$, $0.24 \mu\text{g m}^{-3}$, $1.14 \mu\text{g m}^{-3}$ and $4.79 \mu\text{g m}^{-3}$ in Huaibei, Hefei, Wuxi and Jinhua cities, respectively. Meteorological conditions have a greater effect on O₃ than on PM_{2.5}, varying with city and time (Supplementary Figure S6).

3.3.2 Meteorological-driven trends of PM_{2.5} and O₃

PM_{2.5} has a positive correlation with O₃ at high temperatures and a negative correlation at low temperatures (Zhang et al., 2018; Chen et al., 2019; Yang et al., 2021). In general, the synergic pollution of particulate matter and O₃ occurs in April and October (Supplementary Figures S3, S4).

Figure 10A shows the annual and monthly trends of PM_{2.5} concentrations from 2015 through 2021. The mean PM_{2.5} trend observed in Huaibei, Hefei, Wuxi, and Jinhua cities were $-1.7 \mu\text{g m}^{-3} \text{yr}^{-1}$, $-4.6 \mu\text{g m}^{-3} \text{yr}^{-1}$, $-4.2 \mu\text{g m}^{-3} \text{yr}^{-1}$, and $-3.8 \mu\text{g m}^{-3} \text{yr}^{-1}$, with meteorological factor contributions of $0.05 \mu\text{g m}^{-3} \text{yr}^{-1}$, $0.05 \mu\text{g m}^{-3} \text{yr}^{-1}$, $0.11 \mu\text{g m}^{-3} \text{yr}^{-1}$, and $-0.01 \mu\text{g m}^{-3} \text{yr}^{-1}$, respectively. The changes in meteorological conditions caused an increase or decrease of particulate matter, and their relative contributions for April and October are estimated to be 21% and -21% in Huaibei, 11% and -12% in Hefei, 20% and 3% in Wuxi, and -5% and 21% in Jinhua. We further identified the most important meteorological factors that affect the long-term PM_{2.5} trend. For example, in Huaibei, the 2-m relative humidity (RH₂, $-1.35\% \text{yr}^{-1}$) was responsible for 49% of total meteorological contributions in April, and the planetary boundary layer height (PBLH, -6.8m yr^{-1}) was responsible

for 34% in October. As for Hefei, the primary meteorological factor to drive the long-term PM_{2.5} trend was the daytime PBLH (-12.29 m yr^{-1}), which accounted for 45% of meteorological contributions in October, while sea level pressure (SLP, 0.77 hPa yr^{-1}) was the primary factor in April. As for Wuxi, the most important meteorological factor was SLP (0.68 hPa yr^{-1}) accounting for 61% of meteorological contributions in April, while daytime total cloud cover (TCC, $-0.03\% \text{ yr}^{-1}$) accounted for 31% of meteorological contributions in October. For Jinhua, the precipitation changes (-0.03 mm yr^{-1}) could explain 43% of the meteorological-driven PM_{2.5} trend in April, and the daytime PBLH (15.07 m yr^{-1}) could explain 56% of the change in October.

Supplementary Figures S7A,B present PM_{2.5} changes relative to the previous year (increment) for April and October. SLP and RH play prominent roles in the PM_{2.5} increment in April when meteorological impacts outweigh non-meteorological effects, whereas PBLH is prominent in October. According to Chen et al. (2020), the most significant changes in favorable climatic circumstances for improving PM_{2.5} air quality are lowering RH₂ and deepening PBLH. The deep boundary layer can enhance the diffusion of pollutants through turbulent transport and vertical mixing, which reduces PM_{2.5} pollution (Liu et al., 2018; Su et al., 2018; Miao et al., 2019). Particularly in periods of heavy winter pollution, the formation of secondary particles is inhibited by a reduction in water vapor, which ultimately results in a reduction in PM_{2.5} (Song et al., 2018). On the other hand, high RH promotes aerosol hygroscopic growth and hastens gaseous pollutant transformation into secondary aerosol components (Cheng et al., 2015; Qiao et al., 2016).

Figure 10B displays the growth rates of yearly and monthly MDA8 O₃ from 2015 through 2021. The mean growth trends of MDA8 O₃ were $+2.42 \mu\text{g m}^{-3}\text{yr}^{-1}$, $+4.87 \mu\text{g m}^{-3}\text{yr}^{-1}$, $+2.7 \mu\text{g m}^{-3}\text{yr}^{-1}$, and $+0.83 \mu\text{g m}^{-3}\text{yr}^{-1}$ in Huaibei, Hefei, Wuxi, and Jinhua cities, respectively, in which the contributions from meteorology were $-0.3 \mu\text{g m}^{-3}\text{yr}^{-1}$ (-12%), $+0.26 \mu\text{g m}^{-3}\text{yr}^{-1}$ (5%), $+0.34 \mu\text{g m}^{-3}\text{yr}^{-1}$ (12%), and $+0.43 \mu\text{g m}^{-3}\text{yr}^{-1}$ (52%). The non-meteorological contributions are positive for all cities. The fact that the non-meteorological contribution in Hefei was significantly more than the meteorological contribution suggested that anthropogenic activity was directly responsible for worsening O₃ pollution. The meteorology contributions to MDA8 O₃ growth trends in April and October are estimated to be 65% and -4% for Huaibei, 7% and 35% for Hefei, 41% and 68% for Wuxi, and 64% and 82% for Jinhua. Further investigation was conducted to identify the most important meteorological factors to O₃ changes. The daytime surface solar radiation (SSRD) played the most important role in meteorologically induced MDA8 O₃ changes ($-0.2 \text{ W m}^{-2} \text{ yr}^{-1}$), accounting for 46% of the change in April in Huaibei, while the SLP (0.41 hPa yr^{-1}) accounted for 37% in October. The primary meteorological factor was total precipitation (-0.02 mm yr^{-1}),

accounting for 44% of meteorological contributions for Hefei in April, and TCC ($-0.01\% \text{ yr}^{-1}$) 26% of meteorological contributions in October. As for Wuxi, the daytime SSRD ($0.21 \text{ W m}^{-2} \text{ yr}^{-1}$) could explain 39% and 43% of the total meteorological contributions for April and October, respectively. As for Jinhua, the most significant meteorological factor was precipitation (-0.03 mm yr^{-1}), attributing to 52% in April, and daytime SSRD ($0.13 \text{ W m}^{-2} \text{ yr}^{-1}$) for 42% in October.

Supplementary Figures S7C, D show the meteorological driven MDA8 O₃ changes relative to the previous year for April and October. Solar radiation and cloud cover are the major meteorological factors that impact O₃ concentrations in April and October. Previous studies have shown a direct positive correlation between temperature and O₃, that is, higher temperature accelerates biological emissions of precursors and chemical reaction rate and, in turn, promotes O₃ production (Aw and Kleeman 2003; Gupta and Mohan 2015). Similarly, intense solar radiation accelerates chemical reactions and raises O₃ levels (Chang et al., 2019). In addition to the reduced downward ultraviolet radiation on the ground, through aqueous phase chemistry and photochemistry, clouds can diminish O₃ by improving oxidant elimination and lowering tropospheric oxidation capacity (Lehveleveld and Crutzen 1990). Low RH₂ increases O₃ because certain complicated chemical reactions are inhibited at high humidity levels. Furthermore, low RH₂ is always accompanied by less cloudiness to speed up the photochemical synthesis of O₃ (Camalier et al., 2007). O₃ can also be significantly impacted by 500 hPa winds, for example, in the case of Huaibei in October 2020 and 2021, because wind fields have the potential to significantly affect O₃ and its precursors through transportation (Lu et al., 2019; Liu and Wang 2020).

4 Conclusion

This study investigated the spatio-temporal characteristics of urban PM_{2.5} and O₃ pollution in the Yangtze River Delta, and developed stepwise multiple linear regression models to quantify meteorological and non-meteorological contributions to pollution. In light of spatial heterogeneity, the four city clusters are classified and employed to compare uni-PM_{2.5}, uni-O₃, and bi-PM_{2.5}-O₃ at a regional scale. From 2015 to 2021, PM_{2.5} declines, but MDA8 O₃ rises gradually at different rates in the four city clusters. The uni-PM_{2.5} mainly occurs in winter and decreases from northwest to southeast. The uni-O₃ mainly occurs in warm times from April to October and decreases from northeast to southwest. The bi-PM_{2.5}-O₃ usually appears in April and October and covers central and southern Jiangsu province. The contribution of non-meteorological factors to pollution changes is far greater than meteorological factors.

We also found that PM_{2.5} links closely with O₃ in a synergistic manner in YRD. That is, the higher the photochemical activity levels, the larger the proportion of

secondary components in PM_{2.5}. And the increase of PM_{2.5} concentration in these cities can promote O₃ production to some extent in April and October. Meteorological conditions have a greater effect on O₃ than on PM_{2.5}, varying with city and time. Among them, the contributions of non-meteorological changes to PM_{2.5} decrease were more than 90% of the total decrease in four cities. However, the impact of meteorological conditions on PM_{2.5} outweighed that of anthropogenic emissions in specific years. Therefore, the inter-annual variability in meteorology must be considered in designing future control strategies to improve air quality. SLP and RH play prominent roles in the PM_{2.5} increment in April when meteorological impacts outweigh non-meteorological effects, whereas PBLH is prominent in October. Solar radiation and cloud cover are the major meteorological factors that impact O₃ concentrations in April and October. Key meteorological factors vary by location and time, and should be taken into account in future more refined pollution control.

It is evident from the findings above that the effects of on- and non-meteorological factors on particle and O₃ pollution vary from time to place. Determining the roles of natural and anthropogenic factors will help us to formulate future prevention and control policies to air pollution better aiming at one city or one region. It should be noted that the real contributions of natural and anthropogenic changes cannot be ideally differentiated due to the complexity of atmospheric processes. Therefore, it is necessary to accurately evaluate the contribution of meteorology and corresponding atmospheric processes to air pollution in the future.

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 authors.

Author contributions

QZ and YY conceived of the presented idea. QZ developed the theory and performed the computations. HG and YW verified the

analytical methods. HW and WW encouraged QZ to investigate and supervised the findings of this work. BX and TC confirmed the conceptualization and proof-reading. All authors discussed the results and contributed to the final manuscript.

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Conflict of interest

The authors declare that the research was conducted in the absence of any commercial or financial relationships that could be construed as a potential conflict of interest.

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Supplementary material

The Supplementary Material for this article can be found online at: <https://www.frontiersin.org/articles/10.3389/fenvs.2022.1104013/full#supplementary-material>

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