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Long-Term Change Analysis of $PM_{2.5}$ and Ozone Pollution in China's Most Polluted Region during 2015–2020

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Abstract: In this study, a time change analysis of fine particulate ($PM_{2.5}$) emission in multi-resolution emission inventory in China (MEIC) from 2013 to 2016 was conducted. It was found that $PM_{2.5}$ emissions showed a decreasing trend year by year, and that the annual total emission of $PM_{2.5}$ decreased by 28.5% in 2016 compared with that of 2013. When comparing the observation data of $PM_{2.5}$ and ozone (O_3), it was found that both $PM_{2.5}$ and O_3 show obvious seasonal changes. The emission of $PM_{2.5}$ in autumn and winter is higher than that in summer, while that of O_3 is not. Our study showed that in the 2015–2020 period, annual mean concentrations of $PM_{2.5}$ and O_3 in Beijing varied from 80.87 to 38.31 μg m^{-3} and 110.75 to 106.18 μg m^{-3}, respectively. Since 2015, the observed value of $PM_{2.5}$ has shown an obvious downward trend. Compared with 2015, the average annual $PM_{2.5}$ concentrations in Beijing, Shanghai, Xuzhou, Zhengzhou, and Hefei in 2020 had decreased by 52.62%, 40.35%, 22.2%, 46.84%, and 45.11%, respectively, while O_3 showed an upward trend. Compared with the annual averages of 2015 and 2020, Beijing and Shanghai saw a decrease of 4.13% and 8.46%, respectively, while Xuzhou, Zhengzhou, and Hefei saw an increase of 7.08%, 19.46%, and 41.57%, respectively. The comparison shows that $PM_{2.5}$ is becoming less threatening in China and that ozone is becoming more difficult to control. Air pollution is a modifiable risk factor. Appropriate sustainable control policies are recommended to protect public health.

Keywords: MEIC; $PM_{2.5}$; time variation; O_3

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1. Introduction

In the past three decades, with the rapid growth of China's economy and the rapid advancement of industrialization and urbanization, China's environmental pollution problems have become more and more serious. Among them, air pollution is particularly noticeable. Air compound pollution creates serious environment and health problems in urban areas of China [1,2]. As representative pollutants of air compound pollution, O_3 and $PM_{2.5}$ in the ambient atmosphere are becoming a pervasive air quality problem facing China [3–5]. According to a report on the ecological environment condition in China in 2017, with regard to statistics in 2017, of the more than 338 cities in China there are 99 cities with standard ambient air quality, accounting for 29.3% of the total number of cities; and there are 239 with urban environmental air quality, accounting for 70.7%. These 338 cities had 2311 days of high pollution and 802 days of very high pollution, with $PM_{2.5}$ as the primary pollutant (accounting for 74.2 percent of the days with heavy pollution) [6].

$PM_{2.5}$ and O_3 pollution are health threats of extensive concern. The associated health impacts of ambient $PM_{2.5}$ and ozone have been studied comparatively worldwide. $PM_{2.5}$ and O_3 are considered the causes of increased health risks in the U.S. [7–9]. In Europe, Sicard found that the annual $PM_{2.5}$-related death rate decreased by 4.85 per 106 inhabitants between 2000 and 2017, while the ozone-related rate increased by 0.55 per 106 inhabitants [10]. In India, scholars determined that the ozone-related health impact was much lower than the $PM_{2.5}$-related impact [11,12]. In Iran, ozone-related health impacts have been found to be lower than those that are $PM_{2.5}$ related in Tehran, Ahvaz, and Karaj [13–15]. In the U.S. and Europe, ozone pollution has become a principal public health issue. In Asian countries, ozone-related health risks were still significantly lower than those that are $PM_{2.5}$ related, and individual studies even consider them to be negligible. Worldwide studies reveal a need to regulate $PM_{2.5}$ and ozone risks according to risk characteristics and stages. China, the world's most populous country, is also one of the most severely polluted countries by $PM_{2.5}$ and O_3 [16–18]. A two-pollutant health impact study on China can support synergistic control and provide an informative reference for other regions and countries [19].

In order to better grasp the overall situation of pollutant inventory in China and effectively improve air quality, the National Environmental Monitoring Station began to compile the inventory of air pollution sources in 28 cities in China from 2016. Other provinces and cities across the country have also carried out the corresponding inventory compilation work in the hope of understanding the causes and treatment methods of heavy air pollution in China. The purpose of the emission inventory of air pollutants is to estimate the emissions of various air pollutants in a region based on relevant information of emission sources, including important data for both understanding the emission characteristics of regional pollutants and accurately simulating air quality [20]. At the same time, the emission source inventory is also an important basic data source of environmental air quality management, which is the key to solving air pollution [21,22]. Current global emissions inventories mainly include the United Nations Framework Convention on Climate Change (UNFCC) emissions inventory, the emissions inventory of interactions and synergies between greenhouse gases and air pollution (Greenhouse Gas and Air Pollution Interactions and Synergies ((GAINS)) [23–25], the Emission Database for Global Atmospheric Research (EDGAR) and the Global Emissions Initiative (GEIA. They also include some intercontinental inventories, such as the Transport and Chemical Evolution over the Pacific (TRACE-P) [21], Intercontinental Chemical Transport Experiment-Phase B (INTEX-B) [26], MIX for MICS-Asia (Model Inter-Comparison Study for Asia) [27], Hemispheric Transport of Air Pollution (HTAP) [28], and Evaluating the Climate and Air Quality Impacts of Short-Lived Pollutants (ECLIPSE) emission inventories [29]. This study uses the MEIC [30–34]. The MEIC inventory includes provincial emissions and grid emissions data. Emissions data include electric power, industrial, civil, traffic and agriculture industry (as well as another five industries) data, and 0.25, 0.5, and 1.0 degrees of three kinds of grid emissions from a month-to-month inventory of spatial resolution. The inventory can conduct SAPRC99, SAPRC07, CB05, CBIV output, and RADM2′s five chemical mechanisms. In this study, the annual changes in $PM_{2.5}$ emissions in the MEIC inventory were compared and analyzed.

Since 2017, the ministry of environmental protection has formulated targeted pollutant emission reduction plans and heavy pollution weather early warning plans in autumn and winter. Therefore, in the autumn and winter of 2017, the air quality of Beijing, Tianjin, and Hebei and their surrounding areas was significantly improved (although the air quality in the autumn and winter of 2018 was slightly worse than that in 2017, and there has even been heavy pollution over a large area for a long time). However, compared with that before 2017, the air quality of the whole year was significantly improved, which shows that the pollutant emission list is the basis of air pollution control according to the report on the ecological environmental condition in China in 2020. In 2020, among the 337 prefecture-level and above cities in China, the ambient air quality of 202 cities reached the standard and 135 cities exceeded the standard. The exceeding standard ratio has decreased from 78.4% in 2015 to just 40.1%. However, the number of days exceeding the standard with $PM_{2.5}$ as the primary pollutant accounts for 51.0% of the total number [35]. It can be seen that $PM_{2.5}$ is still the main pollutant in China. In these years, the proportion of the number of days exceeding the daily average of ozone in the monitoring days has increased year by year from 4.6% in 2015 to 37.1% in 2020. Promoting the collaborative control of $PM_{2.5}$ and O_3 has become the primary task of the environmental department and the research difficulty of the scientific community. This paper will compare and analyze the emission changes in $PM_{2.5}$ and O_3 in recent years.

2. Study Area and Data

The research area of this study is the central and eastern regions of China, covering most regions (such as the central and eastern parts of China) and also the region with the most serious air pollution in China (Figure 1). The pollutant emission inventory data we use, hosted by Tsinghua university, developing the MEIC inventory model provides the data [36]. The spatial resolution of 0.25 degrees, the extract of $PM_{2.5}$ to scale in 2013–2016, and the annual gross scale were analyzed.

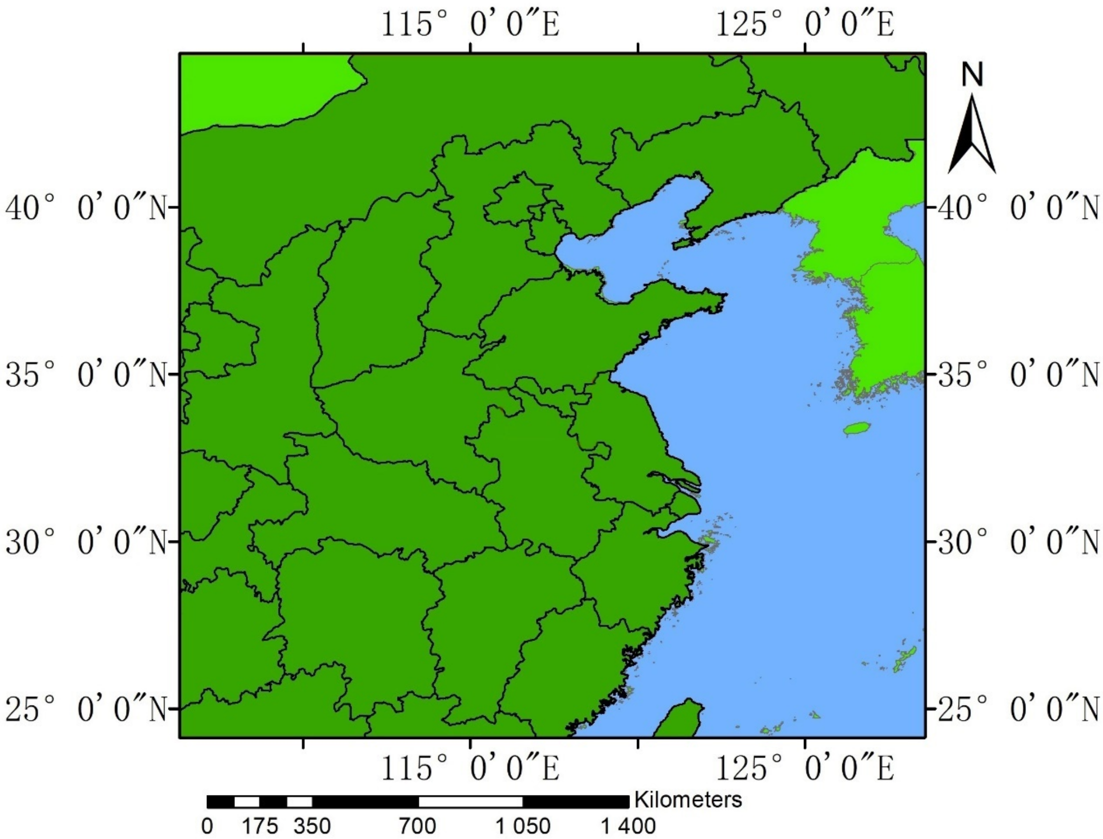

Figure 1. The study area.

To calculate the long-term health impacts at the city level, we use the daily averaged $PM_{2.5}$ concentrations and daily 1-h maximum ozone concentration each year as the primary data. We obtained the hourly concentration data of $PM_{2.5}$ and ozone from May 2015 to December 2020 from the China National Environmental Monitoring Centre air quality real-time publishing platform [37]. This platform mainly monitors the one-hour average concentrations of sulfur dioxide (SO_2), nitrogen dioxide (NO_2), carbon monoxide (CO), $PM_{2.5}$, particulate matter (PM_{10}), O_3 and other pollutants. We extracted the $PM_{2.5}$ and O_3 hourly data from 2014 to 2020 for comparative analysis, in which O_3 has the maximum value over one hour on the daily average scale.

3. Results and Discussion

This section may be divided by subheadings. It should provide a concise and precise description of the experimental results, their interpretation, and the experimental conclusions that can be drawn.

3.1. Change Analysis of Emission Inventory

Figure 2 shows the spatial distribution comparison of the annual total $PM_{2.5}$ emissions from 2013 to 2016 in MEIC, with a spatial resolution of 0.25 degrees. As can be seen from the figure, $PM_{2.5}$ emissions are reasonably distributed in space, mainly in urban areas, north China and the Beijing–Tianjin–Hebei region. On an annual scale, $PM_{2.5}$ emissions show a decreasing trend year by year. Table 1 shows that $PM_{2.5}$ emissions decreased from 11.33 million tons in 2013 to 8.1 million tons in 2016, a decrease rate of 28.5%. Total emissions in 2014 were 9.27 percent lower than in 2013, in 2015 11.27 percent lower than in 2014, and in 2016 11.22 percent lower than in 2015. It shows that the national emission reduction measures formulated according to the research results of scientific researchers are effective.

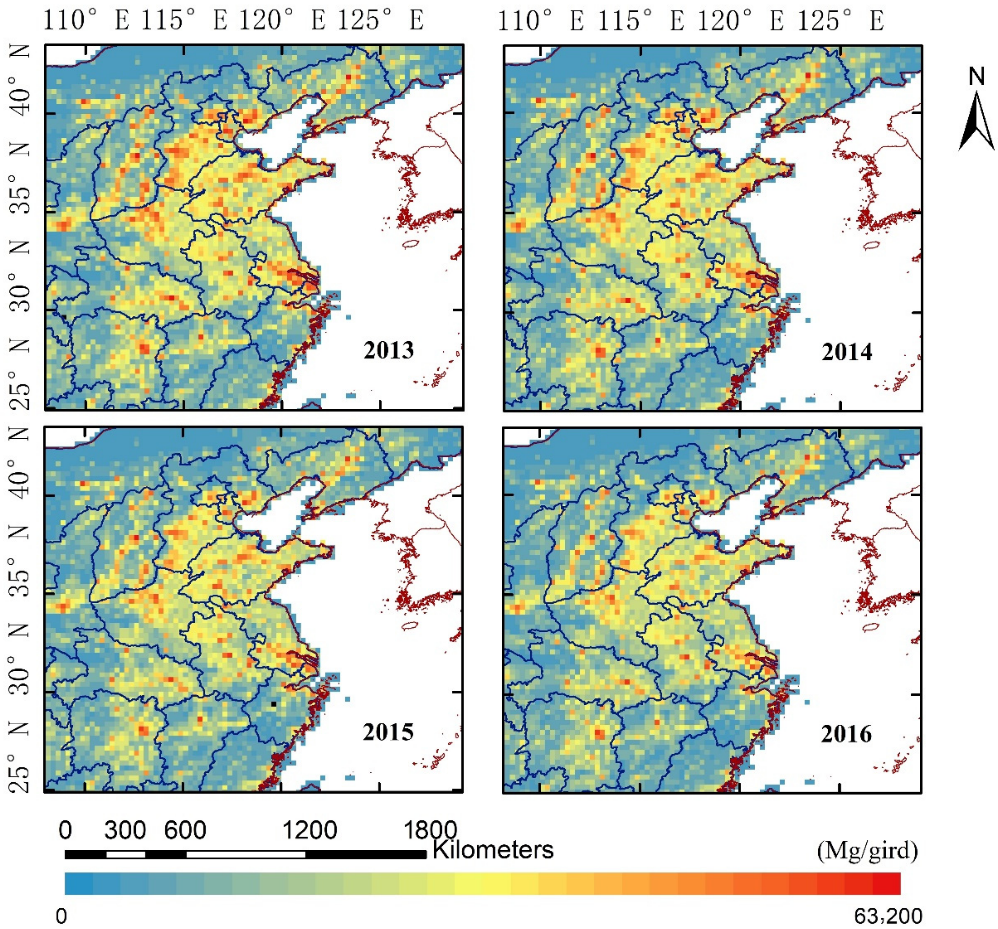

Figure 2. Spatial distribution of $PM_{2.5}$ emission (2013–2016).

Table 1. $PM_{2.5}$ annual total emission data.

Year	$PM_{2.5}$ Emissions (Tons)	Decrease Ratio Compared with the Previous Year
2013	11,333,189.55	-
2014	10,282,426.54	9.27%
2015	9,123,665.07	11.27%
2016	8,100,167.99	11.22%

Figure 3 shows the comparison of spatial distribution changes in $PM_{2.5}$ emission data extracted from April, July, November, and December from 2013 to 2016. The years from top to bottom are 2013, 2014, 2015, and 2016. Changes over the years also show that the total amount of emissions is declining, especially in some key emission areas and metropolitan areas. Figure 4 shows the emission comparison data of each month. The emission value of each month is decreasing year by year, and the annual emission distribution is higher in autumn and winter than in summer. In January and December 2013, the total emission exceeded 1.2 million tons, while in summer in July, it was around 800,000 tons. By 2016, emissions fell below 1 million tons in both January and December, and below 600,000 tons in July.

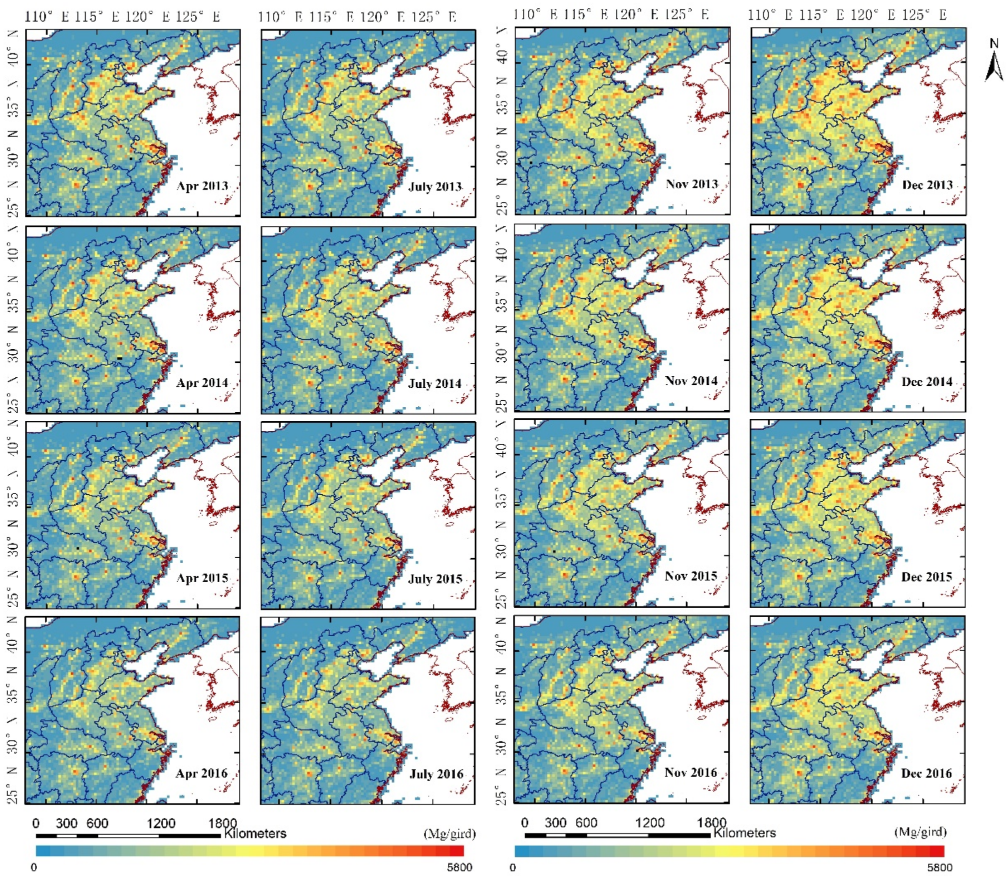

Figure 3. Spatial distribution of monthly total emission of $PM_{2.5}$ (2013–2016).

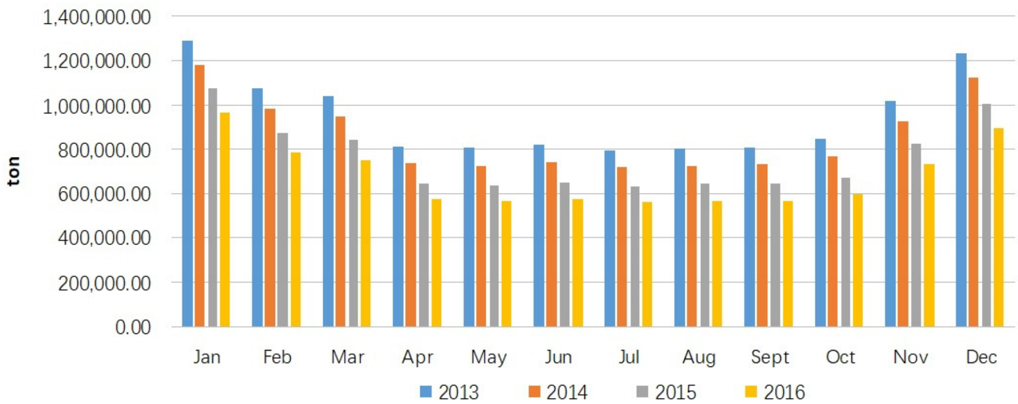

Figure 4. Comparison of total $PM_{2.5}$ emissions in MEIC (2013–2016).

3.2. *Analysis of Long-Term Change in $PM_{2.5}$ and O_3 Concentration Data*

Figures 5 and 6 compare the monthly mean spatial distribution of the $PM_{2.5}$ observed concentrations at each station from April and July, as well as November and December, in 2015 to 2018, respectively.

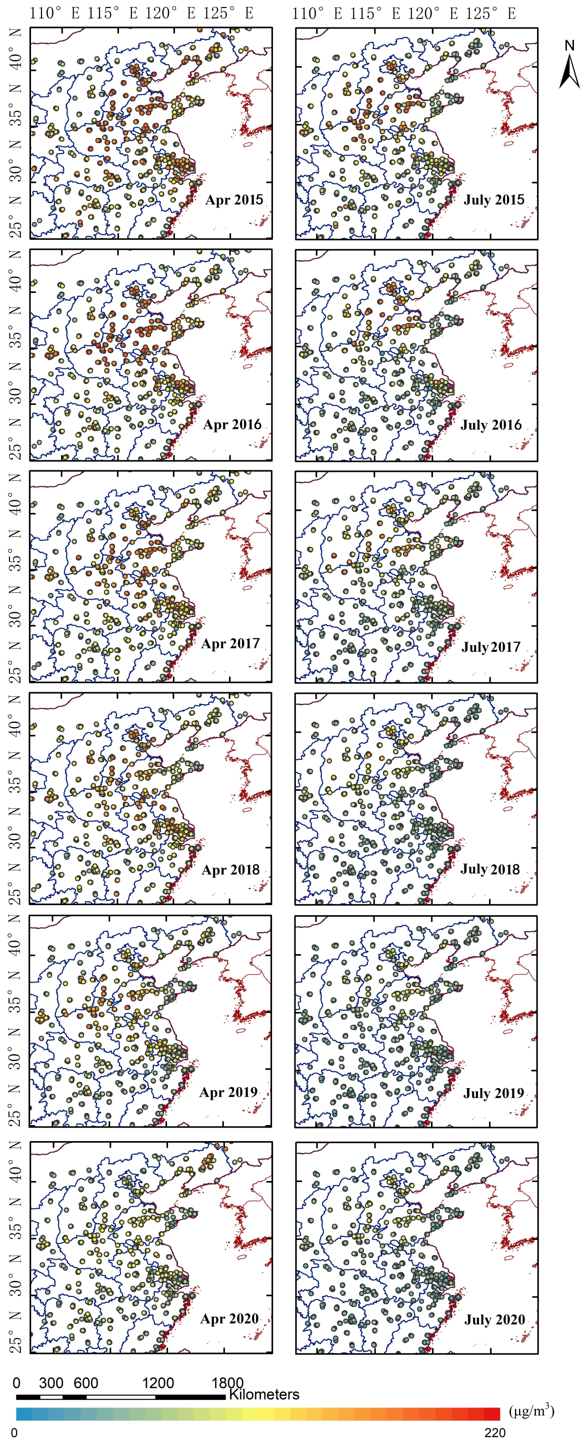

Figure 5. $PM_{2.5}$ concentration values observed in April and July (2015–2020).

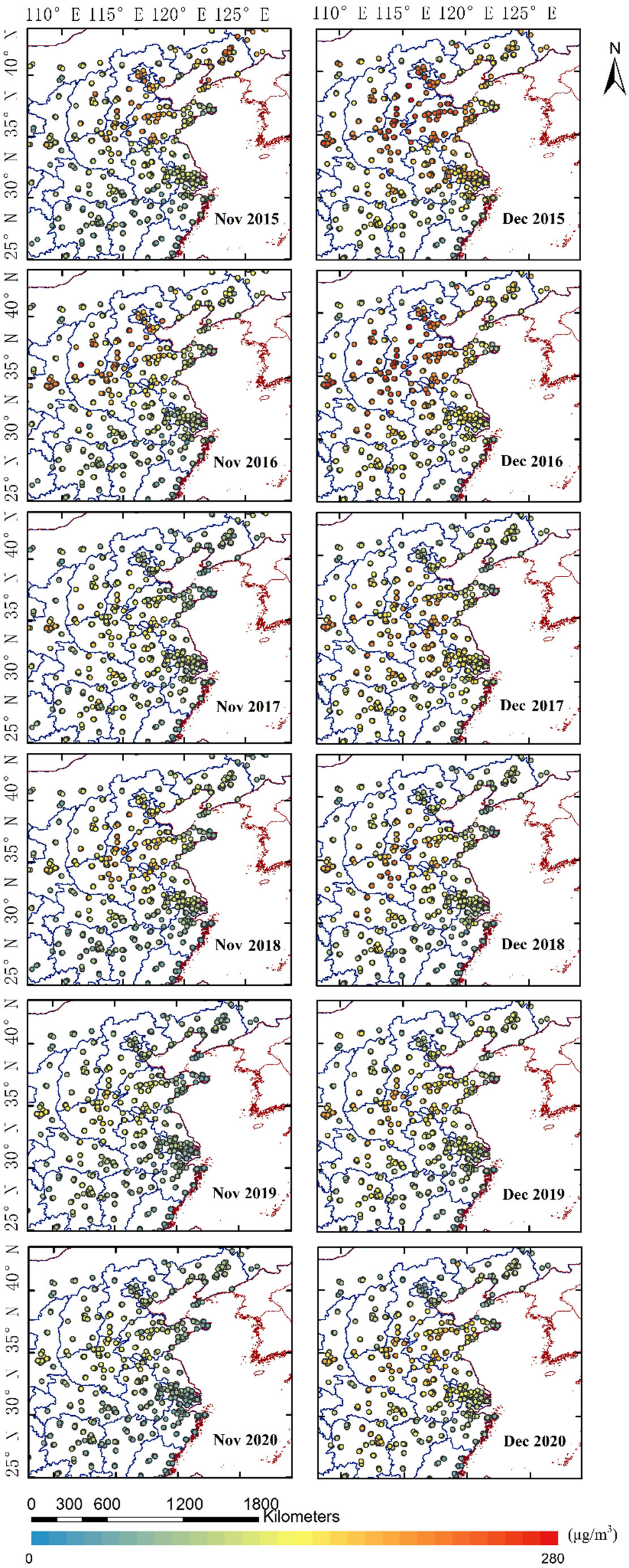

Figure 6. $PM_{2.5}$ concentration values observed in November and December (2015–2020).

Figures 7 and 8 show the comparison diagrams of O_3. The figures from top to bottom show the data of stations in the corresponding months of 2015, 2016, 2017, and 2018, respectively. It can be seen from the figure that the monthly average concentration of $PM_{2.5}$ has been decreasing since 2017. In particular, in the central and eastern parts of China and north China, the concentration of $PM_{2.5}$ has decreased significantly, which corresponds to the emissions in the MEIC inventory. This indicates that, from 2017, the role of a series of national air pollution control measures has begun to appear. However, the observed value of ozone did not decrease, and there is a rising trend. Figure 9 is the comparison of the growth rate (GR) between the monthly average of 2020 and the monthly average of 2015 for all sites (valid data sites). Figure a shows the comparison of the monthly average growth rate of $PM_{2.5}$ for all sites in April, July, November, and December 2020 and April, July, November, and December 2015. The number of sites whose growth rate exceeds 50% accounts for 4% of the total number of sites, and the growth rates between 20 and 50%, between 0 and 20%, between −10 and 0%, between −50 and −10%, and between −100 and −50% are 3%, 8%, 8%, 68%, and 9% respectively. From the comparison of the four months, it can be found that the monthly average emission of $PM_{2.5}$ has decreased significantly at the site scale and the monthly average emission of at least 70% of the sites has decreased. However, O_3 is a different situation. O_3 emissions generally show an increasing trend. Except for in July, more than 70% of the monthly averages of sites increased in other months. It can be seen that ozone will become the key to air pollution control in the future.

This paper also extracted the effective urban $PM_{2.5}$ daily mean and O_3 daily mean data of Beijing, Shanghai, Xuzhou, Zhengzhou, and Hefei from 13 May 2014 to 31 December 2020. Figure 10 shows the generated daily mean curve comparison diagram of the five cities. Each inverted curve in the figures is the daily mean value of O_3 in the corresponding city. It can be seen from the comparison figures that the daily mean values of $PM_{2.5}$ and O_3 in the five cities show obvious seasonal changes. In winter, $PM_{2.5}$ values are significantly higher than those in summer. The daily mean value of $PM_{2.5}$ in Beijing showed an increasing trend from 2014 to 2016. Since 2017, there has been an obvious downward trend. The situation of the other four cities is basically similar to that of Beijing, but the decreasing range is slightly smaller (among which Xuzhou has the smallest decreasing range). Contrary to the seasonal variation of $PM_{2.5}$, the value of O_3 is higher in summer than in winter under the influence of strong solar radiation and high temperature and the output is stable throughout the year. Figure 11 shows the comparison of the monthly mean values of five cities. It can be seen from the figures that, before 2016, the monthly mean values of $PM_{2.5}$ in the winter in Zhengzhou and Beijing ranked first and second among the five cities, but from 2017, Zhengzhou still ranked first, while Xuzhou also rose to the top two. In January 2018, the monthly average was the first in five cities. However, the overall emission of $PM_{2.5}$ in the five cities showed a downward trend while the emission of O_3 did not. In this work, 24 h and 1 h average concentrations of $PM_{2.5}$ and O_3 for the city were classified into predefined air quality categories based on the WHO's air quality guidelines and interim target levels (namely low pollution (<25 μg m^{-3}), moderate pollution (25–37.5 μg m^{-3}), high pollution (37.5–50 μg m^{-3}), and very high pollution (>50 μg m^{-3}) for PM $_{2.5}$ and low pollution (<100 μg m^{-3}), moderate pollution (100–160 μg m^{-3}), high pollution (160–240 μg m^{-3}), and very high pollution (>240 μg m^{-3}) for O_3). Figure 12 illustrates the temporal distribution of $PM_{2.5}$ and O_3 concentrations separated in the mentioned categories over the study period. Regarding $PM_{2.5}$, the daily concentrations were about 12–43% for all days in the low pollution category over this time period. Overall, the number of days in the low pollution category has increased steadily since 2014, whereas the figure for days with high and very high pollution categories shows the opposite trend. Table 2 shows the comparison data of $PM_{2.5}$ concentration in five cities in autumn and winter from 2014 to 2020, which can clearly reflect the changes in $PM_{2.5}$ and O_3 concentration in each city.

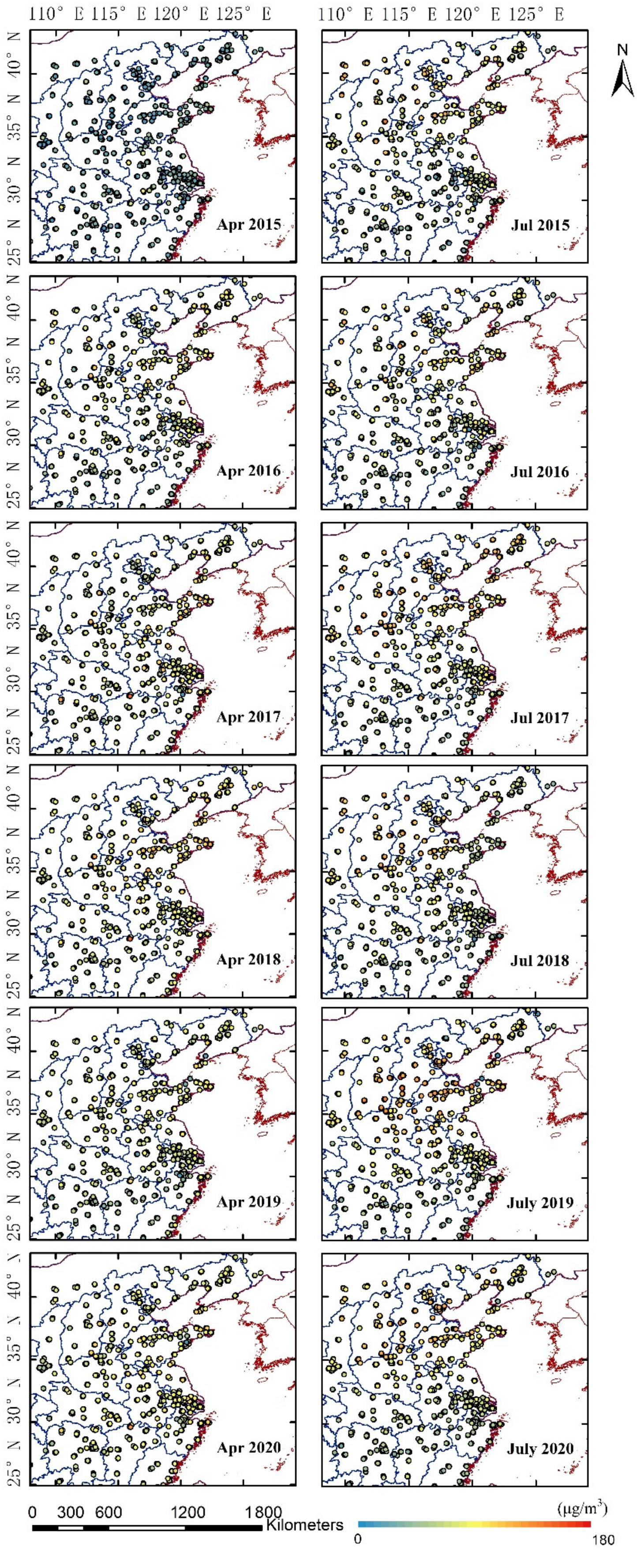

Figure 7. O_3 concentration values observed in April and July (2015–2020).

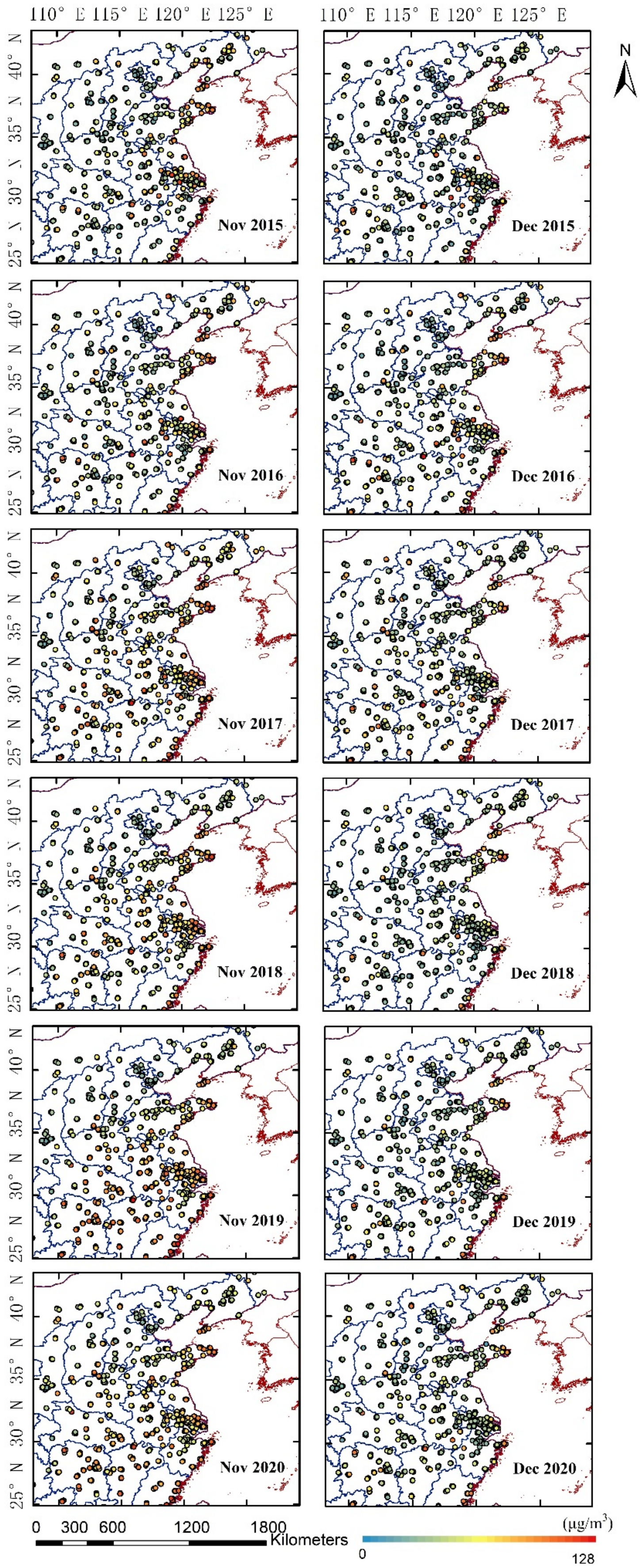

Figure 8. O_3 concentration values observed in November and December (2015–2020).

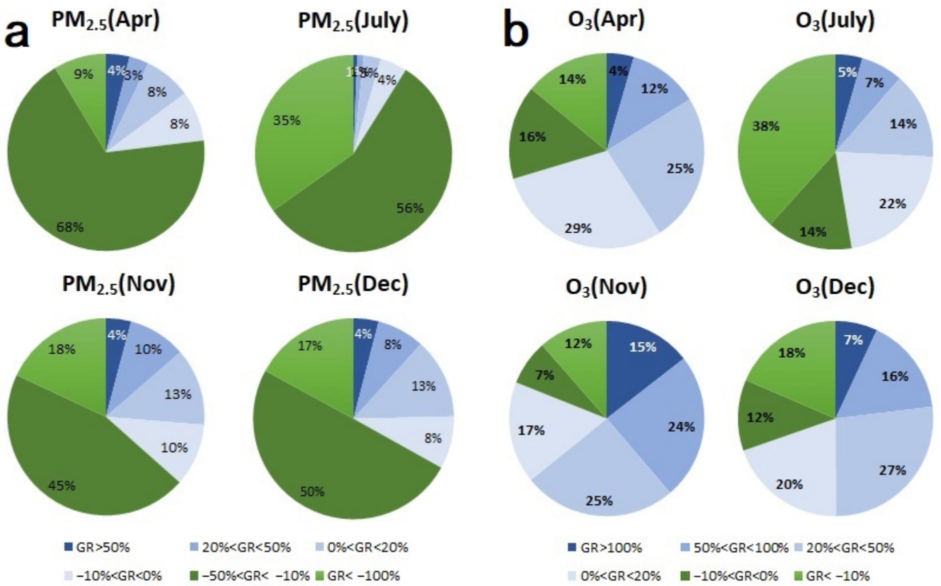

Figure 9. Comparison of monthly average concentration value growth rate of (**b**) O_3 and (**a**) $PM_{2.5}$ at sites.

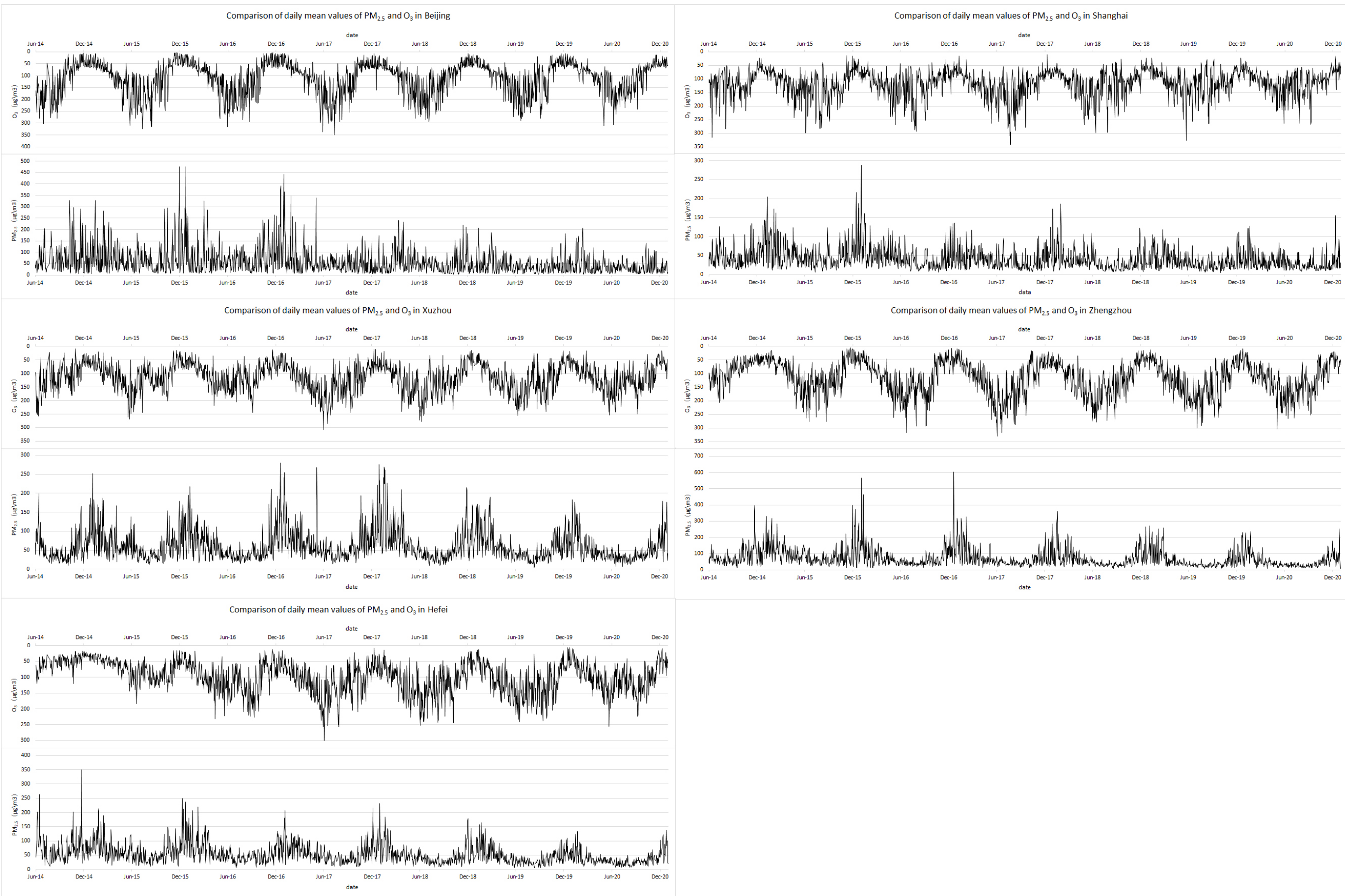

Figure 10. Comparison of the daily mean of urban $PM_{2.5}$ and O_3 (2014–2020).

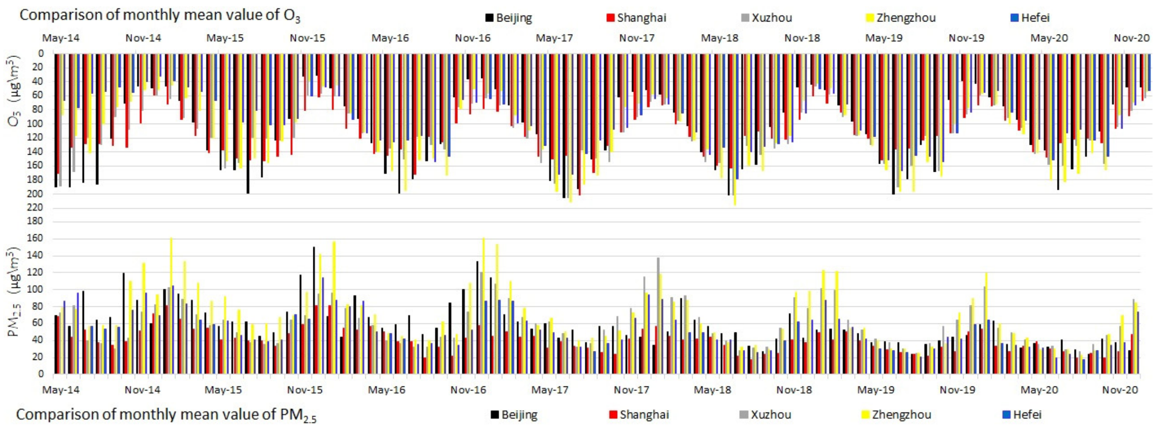

Figure 11. Comparison of the monthly mean of urban $PM_{2.5}$ and O_3 (2014–2020).

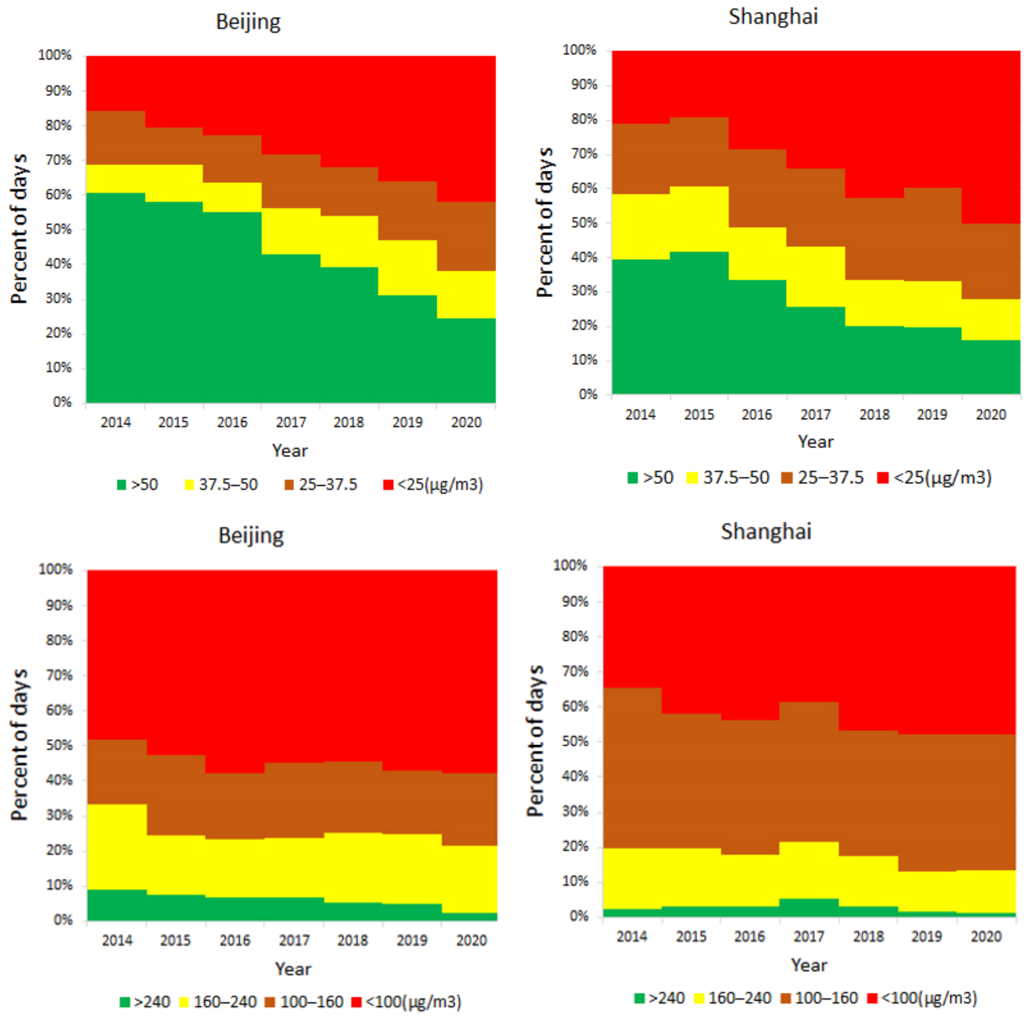

Figure 12. Temporal distribution of $PM_{2.5}$ (above) and O_3 (below) in different categories in Beijing and Shanghai over the study period.

Table 2. Annual mean data of $PM_{2.5}$ and O_3.

	$PM_{2.5}$ ($\mu g\ m^{-3}$)					O_3($\mu g\ m^{-3}$)				
Year	**Beijing**	**Shanghai**	**Xuzhou**	**Zhengzhou**	**Hefei**	**Beijing**	**Shanghai**	**Xuzhou**	**Zhengzhou**	**Hefei**
2015	80.87	53.18	64.27	95.92	65.82	110.75	120.32	107.89	102.94	72.08
2016	72.81	45.64	59.93	78.47	57.19	108.29	117.31	106.30	116.68	104.32
2017	58.02	38.57	68.22	72.02	56.51	113.91	127.17	129.40	124.66	114.63
2018	50.67	35.67	65.92	65.30	48.17	112.53	115.10	122.30	123.92	115.35
2019	42.36	35.24	57.62	59.09	44.10	109.33	110.71	120.07	125.15	114.21
2020	38.31	31.72	50.00	50.97	36.13	106.18	110.14	115.53	122.98	102.05

4. Conclusions

This paper collected the data of monthly and annual $PM_{2.5}$ emissions from the MEIC inventory from 2013 to 2016 and conducted a comparative analysis of the results. It was found that $PM_{2.5}$ emissions showed a decreasing trend year by year. In 2016, compared with 2013, the annual $PM_{2.5}$ emissions decreased by 28.5%, and the average annual decrease was about 10%. Meanwhile, hourly concentration observation data of $PM_{2.5}$ and O_3 at national monitoring stations from 2015 to 2020 were collected in this paper, and then the observation data of $PM_{2.5}$ and O_3 were compared and analyzed. We found opposite trends of $PM_{2.5}$ and ozone in the past six years. $PM_{2.5}$ and O_3 show obvious seasonal changes, and $PM_{2.5}$ emissions are higher in autumn and winter than in summer, while O_3 emissions are higher in summer than in winter. Since 2015, $PM_{2.5}$ observation data and inventory data also showed an obvious downward trend. The average annual $PM_{2.5}$ concentration in Beijing, Shanghai, Xuzhou, Zhengzhou, and Hefei decreased by 52.62%, 40.35%, 22.2%, 46.84%, and 45.11%, respectively, in 2020 compared with 2015. However, O_3 showed an upward trend. Compared with the annual mean value of 2020 and 2015, the annual mean value of O_3 decreased by 4.13% and 8.46% in Beijing and Shanghai, respectively, and increased by 7.08%, 19.46%, and 41.57% in Xuzhou, Zhengzhou, and Hefei, respectively. On the other hand, using the monitoring data in 2020 compared with 2015, the monthly average concentration of $PM_{2.5}$ monitored by more than 70% of the sites has decreased. On the contrary, the monthly average concentration of O_3 monitored by more than 50% of the sites was observed to be rising. Many studies have shown that the reasons for China's $PM_{2.5}$ decline are related to the clean air policies implemented in recent years. For example, Bo Zheng's research pointed out that China's $PM_{2.5}$ emissions were reduced by at least 35% from 2010 to 2017 [34]. The research results suggest that emission control measures are the main drivers of this reduction, in which the pollution controls on power plants and industries are the most effective mitigation measures. These policies mainly include strengthening emission standards in the power and industrial sectors, phasing out outdated industrial capacity, phasing out small high-emitting factories, replacing residential coal use with electricity and natural gas, strengthening vehicle emission standards, retiring old vehicles, and improving fuel quality. However, the decrease in $PM_{2.5}$ also reduces the heterogeneous absorption of HO2 free radicals by aerosols, which in turn intensifies the generation of ozone. Under the current severe atmospheric compound pollution situation, more effective measures are needed to control the emission of nitrogen oxides and volatile organic compounds in order to effectively control ozone pollution. The current air pollution situation in China needs to be tackled now. Otherwise, adverse health effects will accumulate. Air pollution is a modifiable risk factor. The adoption of science-based air quality and emission standards are a key step to reduce the health burden of ambient air pollutions [38]. Appropriate sustainable control policies are needed to protect public health.

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