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Spatiotemporal Characteristics of Air Pollutants (PM_{10}, $PM_{2.5}$, SO_2, NO_2, O_3, and CO) in the Inland Basin City of Chengdu, Southwest China

Kuang Xiao [1,2,*], Yuku Wang [2,*], Guang Wu [1], Bin Fu [2] and Yuanyuan Zhu [2]

[1] Faculty of Geosciences and Environmental Engineering, Southwest Jiaotong University, Chengdu 611756, Sichuan, China; gwu@home.swjtu.edu.cn

[2] Institute of Mountain in Hazards and Environment, Chinese Academy of Sciences, Chengdu 610041, Sichuan, China; fubin32@126.com (B.F.); zhuyuanyuan826@foxmail.com (Y.Z.)

* Correspondence: xiaokuang1983218@sina.com (K.X.); wangyukuan@imde.ac.cn (Y.W.); Tel.: +86-028-8523-0627 (Y.W.)

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Abstract: Most cities in China are experiencing severe air pollution due to rapid economic development and accelerated urbanization. Long-term air pollution data with high temporal and spatial resolutions are needed to support research into physical and chemical processes that affect air quality, and the corresponding health risks. For the first time, data on PM_{10}, $PM_{2.5}$, SO_2, NO_2, O_3 and CO concentrations in 23 ambient air quality automatic monitoring stations and routine meteorological were collected between January 2014 and December 2016 to determine the spatial and temporal variation in these pollutants and influencing factors in Chengdu. The annual mean concentrations of $PM_{2.5}$ and PM_{10} exceeded the standard of Chinese Ambient Air Quality and World Health Organization guidelines standards at all of the stations. The concentrations of PM_{10}, $PM_{2.5}$, SO_2 and CO decreased from 2014 to 2016, and the NO_2 level was stable, whereas the O_3 level increased markedly during this period. The air pollution characteristics in Chengdu showed simultaneously high PM concentrations and O_3. High PM concentrations were mainly observed in the middle region of Chengdu and may have been due to the joint effects of industrial and vehicle emissions. Ozone pollution was mainly due to vehicle emissions in the downtown area, and industry had a more important effect on O_3 in the northern area with fewer vehicles. The concentrations of PM_{10}, $PM_{2.5}$, NO_2 and CO were highest in winter and lowest in summer; the highest SO_2 concentration was also observed in winter and was lowest in autumn, whereas the O_3 concentration peaked in summer. Haze pollution can easily form under the weather conditions of static wind, low temperature and relative humidity, and high surface pressure inside Chengdu. In contrast, severe ozone pollution is often associated with high temperature.

Keywords: air pollutants; exceeding standard levels; spatiotemporal distribution

1. Introduction

During the past 30 years, the rapid urbanization and industrialization in China have greatly affected the ecological environment, and the problem of urban air pollution has become a major focus in recent years [1–4]. Southern China is one of the three areas affected most severely by acid rain, after Western Europe and North America [5,6]. The total sulfur dioxide (SO_2) emissions in China have been decreasing continuously since 2006 [7], but SO_2 emissions are still extremely high in some areas [8]. In addition, haze covers one-quarter of the country's land area and affects 600 million people [9]. In general, air pollution has been improved greatly in recent years, but haze pollution is still a severe environmental problem at present, and is harmful to human health [10–12]. Great attention

has been paid to particulate matter (PM) pollution, including $PM_{2.5}$ (particles with aerodynamic diameter $\leq$ 2.5 μm) and PM_{10} ($\leq$10 μm), but the levels of other pollutants are also growing rapidly, particularly ozone (O_3). According to national monitoring data, O_3 pollution has begun to appear in the summer and has tended to replace $PM_{2.5}$ as the major pollutant in major Chinese cities [13,14]. The combination of winter pollution (PM) and summer pollution (ozone) indicates that China is facing a complex air pollution problem.

Research on air pollution in China included the mechanism of haze formation [15,16], the influence of meteorological and topographic factors on air quality [17,18], temporal and spatial air pollution patterns [19,20], and the long distance transmission of pollutants [21–23]. However, most of these studies are concentrated in the economically developed areas, e.g., the Beijing–Tianjin–Hebei region [24], the Pearl River Delta [25], and the Yangtze River Delta [26], and are dominated by single pollutants (PM or O_3) and short periods (1–2 years of heavy pollution) of study. Few investigations have considered the spatial and temporal variations in various pollutants [27,28].

Chengdu is an important economic center in southwest China and the only mega-city in the west. The area is located on a plain of less than 5000 km^2, with a population of more than 15 million people, as well as 4 million vehicles and a large number of production and service activities [29]. The emission of air pollutants continues at a high level in this area and, due to its unique basin topography, the Chengdu area has been greatly affected by regional haze pollution. Monitoring data over the past three years have shown that O_3 concentrations are increasing year by year, and air pollution is getting worse. However, most previous studies have only considered PM. For example, Liao et al. [17] analyzed the high levels of $PM_{2.5}$ pollution in Chengdu during the winter of 2013, as well as its transmission routes and sources. Huang et al. [30] studied the influence of the meteorological conditions and urban spatial morphology on $PM_{2.5}$ in Chengdu. Zhang et al. [31] analyzed the seasonal pollution characteristics and sources of $PM_{2.5}$ in the urban area by collecting $PM_{2.5}$ samples during typical seasons (2009–2010 years) in Chengdu City. Yang et al. [32] analyzed the factors that influence $PM_{2.5}$ in Chengdu and suggested prevention and control strategies. Few studies have considered the characteristics and causes of the multiple pollutants in air pollution in Chengdu, which may be related to the lack of air quality monitoring data, as only PM_{10}, SO_2, and NO_2 were measured on a regular basis before 30 March 2012. Data on other pollutants such as the hourly and daily concentrations of PM_{10}, $PM_{2.5}$, SO_2, NO_2, O_3 and carbon monoxide (CO) until January 2013 have been released to the public, but the published data only covered eight monitoring sites controlled by the state [33]. Six air pollution parameters (PM_{10}, $PM_{2.5}$, SO_2, NO_2, O_3 and CO) were monitored more comprehensively in real time after January 2015.

Given the rapid change in air pollution, it is necessary to determine the characteristic temporal and spatial changes in pollutants as well as their relationships with meteorological conditions, and to evaluate the changes in air quality to formulate preventative and control measures. In the present study, we conducted continuous monitoring at multiple sites for three years in order to: (1) evaluate the exceeding standard levels and concentrations variations in pollutants in Chengdu; (2) determine the temporal and spatial characteristics of the concentrations of air pollutants; and (3) analyze the relationships between pollutants and meteorological factors.

2. Materials and Methods

2.1. Study Area

Chengdu is the largest city in southwest China, located in the western Sichuan basin on the Chengdu plain hinterland (Figure 1) between 102.9° E to 104.88° E and 30.08° N to 31.43° N, with a total area of 14,605 km^2. There are significant differences in the topography of the city: the northwest is high, the southeast is low, and the difference in height between the east and west is 4966 m. The typical climatic characteristics of the basin are high humidity (annual average relative humidity of 79–84%) and low wind speed (perennial ground wind speed of 0.9–1.4 m/s); this is one of the rare static

wind areas in China. Rainfall is abundant in this area but is distributed unevenly among the seasons, with less rain in the winter and spring compared with summer and autumn, and more than 90% of the annual rainfall is concentrated during June–September. The annual average temperature is about 16 °C and the average number of sunshine hours is less than 1500 h. During 2016, the resident population of Chengdu was 15.918 million, the GDP was 1217.02 billion yuan, and the urbanization rate was 70.6%.

Furthermore, Chengdu has a jurisdiction of over 11 districts, four cities, and four counties. Five districts (Jinjiang, Qingyang, Jinniu, Wuhou and Chenghua) constitute the central city of Chengdu. There are six other districts (Qingbaijiang, Xindu, Pidu, Wenjiang, Longquanyi and Shuangliu) and four cities (Dujiangyan, Qionglai, Chongzhou and Pengzhou), and four counties (Dayi, Jintang, Xinjin and Pujiang) contain satellite cities.

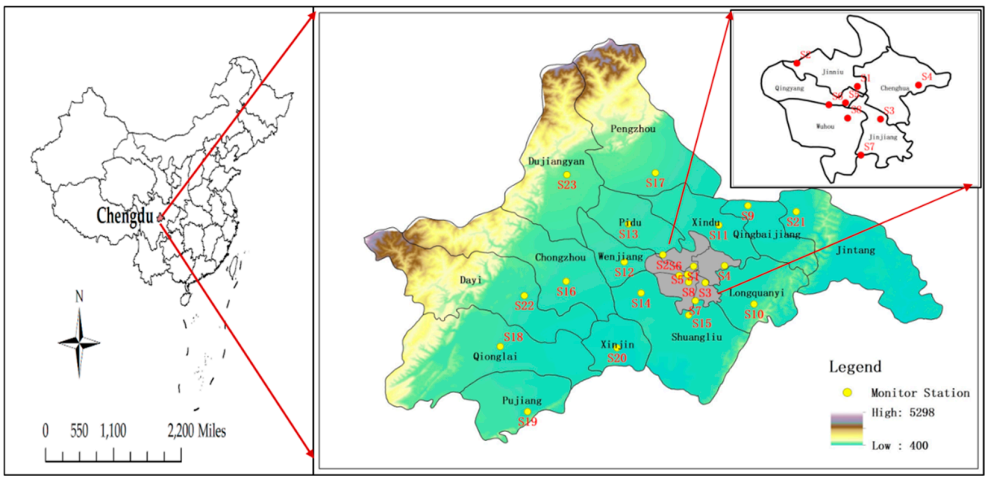

Figure 1. Terrain and locations of the 23 automatic ambient air monitoring stations in Chengdu. The upper right corner of the map shows the central city of Chengdu, and the red dots represent the position of the air monitoring stations.

2.2. Sites, Instruments and Observations

The daily average concentrations of air pollutants (PM_{10}, $PM_{2.5}$, SO_2, NO_2, O_3 and CO) were obtained from 23 automatic ambient air quality monitoring stations in Chengdu (Figure 1). There were eight monitoring stations in the main city, and each satellite city had a monitoring station (except Shuangliu District, which had two). All of the monitoring sites were urban stations, and most sites were located in residential and commercial mixed areas, where the surrounding areas were mostly residential and office areas with roads, and there were no obvious sources of industrial air pollution, so they reflected the environment well in different areas of the city. The heights of the 23 monitoring sites varied, with some on the ground (eight sites) and others on the tops of buildings (15 sites), where the sampling point to ground height ranged from 5 m to 20 m. All sites are in an elevation range between 416 and 683 m.

The instruments used for measuring the concentrations of PM_{10}, $PM_{2.5}$, SO_2, NO_2, O_3 and CO were continuous automatic analysis instruments produced by Thermo Fisher Scientific Inc. (Waltham, MA, USA) and Teledyne API Inc. (San Diego, CA, USA). The PM monitor was used to measure the concentrations of both PM_{10} and $PM_{2.5}$ in ambient air based on light scattering and β-ray attenuation. The SO_2 analyzer employed pulse fluorescence detection to determine the SO_2 concentration in the ambient air. The NO_2 analyzer used chemiluminescence detection to measure the concentration of NO_2 in the ambient air. The O_3 analyzer utilized ultraviolet spectrophotometry to detect the O_3 concentration in the ambient air. The CO analyzer used the gas filter-related infrared absorption method to measure low CO concentrations according to Beer–Lambert's law by comparing the absorption of infrared energy in the sample and a reference gas. The analytical methods used by all of the

instruments and the other performance indicators (e.g., measurement range, minimum detection limit, zero drift and span drift) complied with the technical standards for automated ambient air quality monitoring in China. The ambient air quality automatic monitoring stations were subjected to quality control checks according to Chinese ambient air quality monitoring standards and the validity of the monitoring data was guaranteed.

The observation period was 1 January 2014 to 31 December 2016, a total of 1096 days. A valid annual average of air pollutants at each site was required to be based on at least 324 (88%) valid daily averages in one year; a valid monthly average was required to be based on at least 27 valid daily averages in one month (at least 25 valid daily average concentrations in February); a valid daily averages of air pollutants at each site was required to be based on at least 20 valid hourly averages in one day. O_3 daily average concentrations refer to the daily maximum 8-h mean value in this study, and a valid 8-h running average was required to be based on at least 6 h of concentrations every 8 h. Daily city-wide concentrations were calculated if at least 17 of the 23 sites had valid daily averages; the city-wide annual averages were determined by averaging these concentrations if at least 90% (329 observations) of the city-wide daily averages were valid. All sites met these criteria, and the daily missing data rate of pollutant records for each monitoring site was very low during the study period. It should be explained that sites S19–S22 did not monitor O_3 and CO and S23 did not monitor O_3 between January and December in 2014. Table 1 provides more information on the ambient air quality monitoring stations. Meteorological data from the national basic meteorological station (Wenjiang Station, which is 4 km away from S12) were used for comparisons of the pollutants at S12. The daily meteorological dataset of the station was used in the analysis, including the daily average wind speed (marked as AWs), daily maximum wind speed (MWs), daily mean temperature (T), relative humidity (RH), surface pressure (P), the daily amount of precipitation (R) and sunshine hours (S).

Table 1. Basic details of the 23 ambient air automatic monitoring stations in Chengdu.

Site	Latitude	Longitude	AMSL [1]	SRH [2]	Settings	Administrative	Validity Data Rates (%) [3]					
Code	(°N)	(°E)	(m)	(m)		Divisions	PM_{10}	$PM_{2.5}$	SO_2	NO_2	O_3	CO
S1	30.685	104.0736	509	5	Ground	Jinniu District	98.9	98.9	98.8	98.1	98.3	98.8
S2	30.7236	103.9728	516	15	Roof	Jinniu District	98.4	98.4	98.2	98.4	97.3	98.7
S3	30.6306	104.1122	487	5	Ground	Jinjiang District	99.8	99.3	99.8	99.3	99.3	99.7
S4	30.6872	104.1756	526	15	Roof	Chenghua District	99.5	98.9	99.8	99.5	98.9	99.7
S5	30.6578	104.0539	456	15	Roof	Qingyang District	98.7	99.1	98.8	98.3	98.4	99.2
S6	30.6544	104.0261	450	6	Ground	Qingyang District	99.0	98.6	99.5	99.6	98.6	99.5
S7	30.5706	104.0794	578	15	Roof	Wuhou District	99.3	98.6	99.1	99.3	98.4	99.0
S8	30.6322	104.0575	475	12	Roof	Wuhou District	95.6	94.5	94.7	91.2	94.4	95.0
S9	30.8875	104.2525	475	15	Roof	Qingbaijiang District	97.6	97.4	97.7	97.0	97.0	97.9
S10	30.5589	104.2725	527	7	Ground	Longquanyi District	97.4	98.5	98.5	98.3	97.4	98.8
S11	30.8225	104.1567	449	20	Roof	Xindu District	96.0	96.0	97.9	97.7	97.3	98.7
S12	30.7489	103.86	536	10	Roof	Wenjiang District	97.1	96.8	97.9	97.4	96.4	97.6
S13	30.8631	103.8744	556	18	Roof	Pidu District	97.3	97.9	95.9	97.4	97.2	98.6
S14	30.5958	103.9014	497	8	Ground	Shuangliu District	94.4	94.4	96.3	96.2	95.7	95.1
S15	30.5225	104.0578	471	9	Roof	Shuangliu District	96.7	98.1	97.6	97.6	97.0	99.0
S16	30.6347	103.6547	533	6	Ground	Chongzhou City	97.7	97.9	98.2	98.5	97.4	98.4
S17	30.9969	103.9481	654	15	Roof	Pengzhou City	97.9	98.1	98.2	98.1	97.2	98.2
S18	30.4175	103.4383	500	14	Roof	Qionglai City	98.7	98.7	97.6	99.2	98.1	98.5
S19	30.2006	103.5278	518	10	Roof	Pujiang County	98.7	97.4	98.4	98.5	98.6	98.2
S20	30.4133	103.8217	416	17	Roof	Xinjin County	97.0	96.5	97.2	97.2	96.2	96.9
S21	30.8672	104.4114	478	19	Roof	Jintang County	97.7	97.8	97.0	97.6	96.9	98.2
S22	30.5867	103.62	547	15	Roof	Dayi County	98.9	99.0	99.5	99.6	99.2	99.7
S23	30.9908	103.6575	683	20	Roof	Dujiangyan City	99.9	99.9	99.9	99.9	99.7	99.7

[1] Above mean sea level; [2] Sampling relative altitude; [3] The O_3 observation period of S19–S23 was the same as that of the CO observation period of S19–S22: January 2015 to December 2016 (i.e., 731 days).

2.3. Methods

The meteorological factors affecting the daily variation in pollutant concentrations were evaluated by Pearson correlation analyses with a two-tailed Student's *t*-test. The coefficient of variance was used to assess the homogeneity of the annual average concentrations of different pollutants. Spatial distributions of PM_{10}, $PM_{2.5}$, SO_2, NO_2, O_3 and CO were obtained by the Inverse Distance Weighted interpolation method.

Due to the skewness of the daily data, non-parametric statistics were used, e.g., the single-sample K–S test and Mann–Whitney test. Seasonal average statistics were defined for the winter (December–February), spring (March–May), summer (June–August) and autumn (September–November).

Statistics on the exceeding standard of pollutants were according to the Chinese Ambient Air Quality Standards (CAAQS) and the WHO guidelines (Table 2), respectively.

Table 2. Air pollutant standards for urban areas set by China and the World Health Organization (WHO) (the units for PM_{10}, $PM_{2.5}$, SO_2, NO_2 and O_3 are μg m^{-3}, whereas that for CO is mg m^{-3}).

Items	Average Time	China's NAAQS-2012 [1]	WHO [2] Guideline
PM_{10}	Daily	150	50
	Annual	70	20
$PM_{2.5}$	Daily	75	25
	Annual	35	10
SO_2	Daily	150	20
	Annual	60	-
NO_2	Daily	80	-
	Annual	40	40
O_3	8-h	160	100
CO	Daily	4	-

[1] MEP (2012a); [2] WHO (2005).

3. Results and Discussion

3.1. Air Pollutants Exceeding Standard Levels

Based on the data obtained from 23 monitoring stations in Chengdu City during the study period, we determined the air pollutants (PM_{10}, $PM_{2.5}$, SO_2, NO_2, O_3 and CO) annual and daily average concentrations city-wide and each station and evaluated the air pollutants exceeding the standard levels.

Figure 2 shows the city-wide annual and daily averaged concentrations of PM_{10}, $PM_{2.5}$, SO_2, NO_2, O_3 and CO over the entire study period, respectively. Specific descriptions are also provided in Figure S1 and Tables S1 and S2, respectively. The average annual concentrations of $PM_{2.5}$ and PM_{10} exceeded the standard for three consecutive years (2014–2016), and were 1.5–2 times those of the CAAQS standards (70 and 35 μg/m^3, respectively), and 5–7 times those of the WHO (20 and 10 μg/m^3, respectively). In addition, the NO_2 annual concentrations were exceeded in 2014 and 2016, which were close to the standards. SO_2 was much lower than the standard. The daily average concentrations of $PM_{2.5}$ was at the "exceedance" level for 337 (30.8%) days during the total study period (1096 days) according to the CAAQS standard, which was the highest of all the pollutants. PM_{10}, O_3 and NO_2 were at the exceedance level for 207 (18.9%) days, 131 (12.0%) days, and nine (0.8%) days, respectively. SO_2 and CO were not at the "exceedance" levels. If we consider the WHO guidelines, $PM_{2.5}$, PM_{10}, SO_2 and O_3 exceedance days reached 1025 (93.5%) days, 972 (88.7%) days, 539 (49.2%) days and 410 (37.4%) days, respectively, an increase of 2, 3.7, 538 and 2.1 times compared with the CAAQS standard. Over the entire study period, the daily average concentrations of PM_{10}, $PM_{2.5}$, SO_2, NO_2, O_3 and CO ranged between 17–426 μg/m^3, 11–323 μg/m^3, 5–61 μg/m^3, 13–95 μg/m^3, 9–248 μg/m^3 and 0.432–2.121 mg/m^3, respectively, and the means and standard deviations were 106 ± 17 μg/m^3, 67 ± 14 μg/m^3, 21 ± 3 μg/m^3, 41 ± 3 μg/m^3, 91 ± 10 μg/m^3 and 1.065 ± 0.075 mg/m^3, respectively.

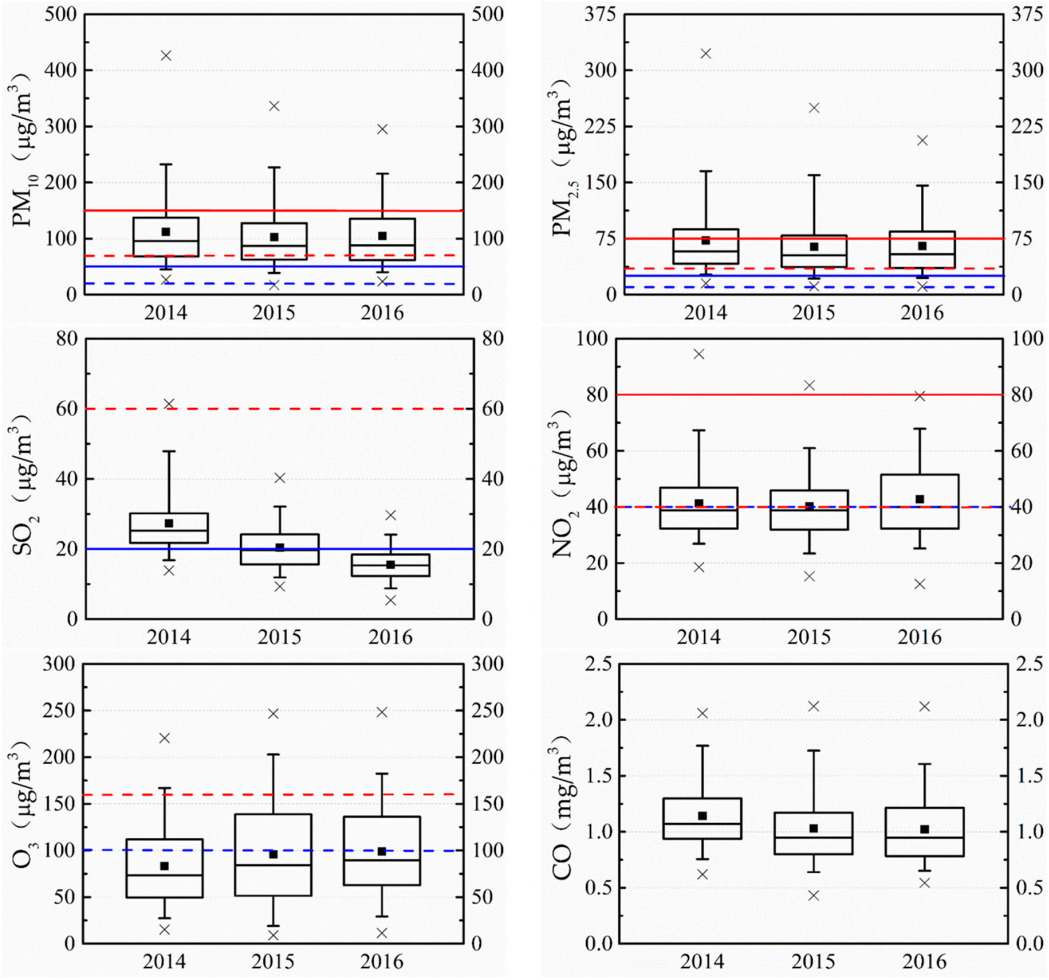

Figure 2. The city-wide average annual and daily concentrations of PM_{10}, $PM_{2.5}$, SO_2, NO_2, O_3 and CO in Chengdu during the study period. The horizontal solid lines indicate the median and the filled squares indicate the mean (i.e., annual average concentrations), and range where the top and bottom of the box indicate the 75th and 25th percentiles, respectively. The top and bottom whiskers indicated the 95th and 5th percentiles, and the forks indicated the maximum and minimum values, respectively. The red solid and dotted lines represent the daily and annual limits specified by CAAQS (Chinese Ambient Air Quality Standards), the blue solid and dotted lines represent the daily and annual limits specified by the WHO.

Annual and daily air pollutant concentrations across the 23 sites are summarized in Tables S1 and S2. The annual average PM_{10}, $PM_{2.5}$, SO_2, NO_2, O_3 and CO concentrations over the entire study period ranged from 77 (S23) to 137 (S8) μg/m^3, 42 (S23) to 89 (S9) μg/m^3, 9 (S19) to 50 (S11) μg/m^3, 20 (S23) to 69 (S1) μg/m^3, 66 (S8) to 118 (S13) μg/m^3 and 0.722 (S23) to 1.487 (S9) mg/m^3, respectively. Obviously, the annual average concentration of PM_{10} and $PM_{2.5}$ at all sites exceeded the standard stipulated by CAAQS and WHO during the study period. The exceeded standard rate of NO_2 annual concentration also reached 45% (31 observations). In addition, the exceeded CAAQS (WHO) frequency of PM_{10}, $PM_{2.5}$, SO_2, NO_2, and O_3 annual concentration at all sites ranged among 9.3–27.7% (66.1–94.5%), 14.8–43.9% (65.5–95.8%), non–excessive (7.8–75.4%), 0–18.0% (no standard), and 4.1–19.6% (31.8–45.8%), respectively. For daily average concentration, the stations with the most exceedance days of PM_{10} included S13 and S2 and $PM_{2.5}$ at S13. PM_{10} exceedance days in S2 reached 299 (27.7%) days; if referring to WHO guidelines, "exceedance" days within the valid monitoring days were 981 (92.0%) days in S13. Standard "exceedance" days of $PM_{2.5}$ daily concentrations at S13 reached 471 (43.9%) (CAAQS) and 1028 (95.8%) (WHO), respectively. S13 was the most seriously

polluted site of all the sites due to O_3, and exceedance days reached 209 (19.6%) (CAAQS) and 487 (45.7%) (WHO). According to the WHO, the most affected site regarding SO_2 pollution was S9, where exceedance days reached 807 (75.4%). S1 had the highest number of NO_2 exceedance days of all the sites, reaching 194 (18.0%).

In terms of the annual and daily concentrations of air pollutants exceeding the standards, $PM_{2.5}$ and PM_{10} were two major pollutants in Chengdu during the study period, similar to most cities in China (e.g., Beijing, Wuhan, and Xi'an) [34–37]. It should be noted that only O_3 exceedance days significantly increased during the study period. The CAAQS SO_2 standard seems more controversial, as the difference between the two standards (CAAQS and WHO) is too large, and the assessment results are also quite different. Therefore, we strongly recommend a revision and update of CAAQS standards for SO_2. In addition, the coefficient variance of PM_{10}, $PM_{2.5}$, SO_2, NO_2, O_3 and CO were 13.4%, 14.2%, 42.8%, 30.8%, 15.6% and 16.8%, respectively. These findings suggested that the daily concentrations of PM_{10}, $PM_{2.5}$, O_3, and CO were more uniform than SO_2 and NO_2.

3.2. Temporal and Spatial Variations in the Air Pollutants

3.2.1. Spatial Pattern and Inter-Annual Variation in Air Pollutants

Spatial and temporal variations in these air pollutants have not been previously reported in Chengdu. The spatial patterns of six pollutants ($PM_{2.5}$, PM_{10}, SO_2, CO, NO_2 and O_3) in the past three years are shown in Figure 3. Additionally, interannual variations of the pollutants concentrations during 2014–2016 are also shown in Figure S2 and Table S1. In terms of the spatial distributions of the air pollutants, the concentrations of PM in the middle of Chengdu, especially areas S2, S4, S8, S9, S11–S14, and S20, were usually higher than those in other regions. The PM_{10} concentration in S14 (126 $\mu g/m^3$) was the highest among the sites and $PM_{2.5}$ was the highest in S13 (81 $\mu g/m^3$), while the S23 site had the lowest PM_{10} and $PM_{2.5}$ concentrations of 80 $\mu g/m^3$ and 45 $\mu g/m^3$, respectively. In general, the spatial distributions of particulate matter pollutants are mainly influenced by local sources. From the interannual variation in particulate matter, the city-wide mean concentrations of $PM_{2.5}$ slightly decreased from 73 $\pm$ 10 $\mu g/m^3$ in 2014 to 65 $\pm$ 8 $\mu g/m^3$ in 2016, which reflected a non-significant decrease in $PM_{2.5}$ concentration over the past years. The interannual variation of PM_{10} was in accordance with that of $PM_{2.5}$. The averaged PM_{10} concentration decreased slightly from 112 $\pm$ 16 $\mu g/m^3$ in 2014 to 105 $\pm$ 12 $\mu g/m^3$ in 2016. Although the PM concentrations as a whole did not display a significant reduction, they showed marked spatial heterogeneity. For example, 15 (4) of 23 sites had a synchronous decrease (increase) in PM_{10} and $PM_{2.5}$ concentrations. The decrease in both PM_{10} and $PM_{2.5}$ concentrations in the middle region of Chengdu (i.e., S3–S10) was the most significant; the decrements averaged 22 $\pm$ 5 $\mu g/m^3$ (ranged 11 to 26 $\mu g/m^3$) and 17 $\pm$ 3 $\mu g/m^3$ (ranged 14 to 25 $\mu g/m^3$), respectively, whereas the average PM in western parts of Chengdu such as S16, S19–S20, and S22 increased and S16 showed the greatest increase in PM_{10} (21 $\mu g/m^3$, 22.3%) and $PM_{2.5}$ (8 $\mu g/m^3$, 12.5%) concentrations.

Higher concentrations of SO_2 were generally observed at sites located in the northern region of Chengdu due to the presence of a large number of industries (such as in S11, S13 and S17) and power plants (such as in S9) (Figure 2). It is noteworthy that the overall SO_2 concentration significantly decreased in recent years, and the mean SO_2 concentrations decreased from 28 $\pm$ 10 $\mu g/m^3$ to 16 $\pm$ 4 $\mu g/m^3$ during the period 2014–2016 (Figure S2, Table S1). The main source of SO_2 was coal-fired boilers (especially coal-fired power plants) in Chengdu. The total industrial SO_2 emissions decreased considerably after the phasing out of small and less efficient coal-fired power generation units and steel plants, the installation of flue gas desulfurization systems in thermal power units, the implementation of strict emissions standards for industrial boilers, the change in fuel from coal to natural gas, and restrictions on the building of new cement plants, ceramics factories, and glassworks. In addition, most sintering processes adopted electrostatic precipitators for PM removal and fabric filters were

more widely used in steel-making, iron-making, and cement production processes, which sharply reduced the emissions of SO_2 [38–40].

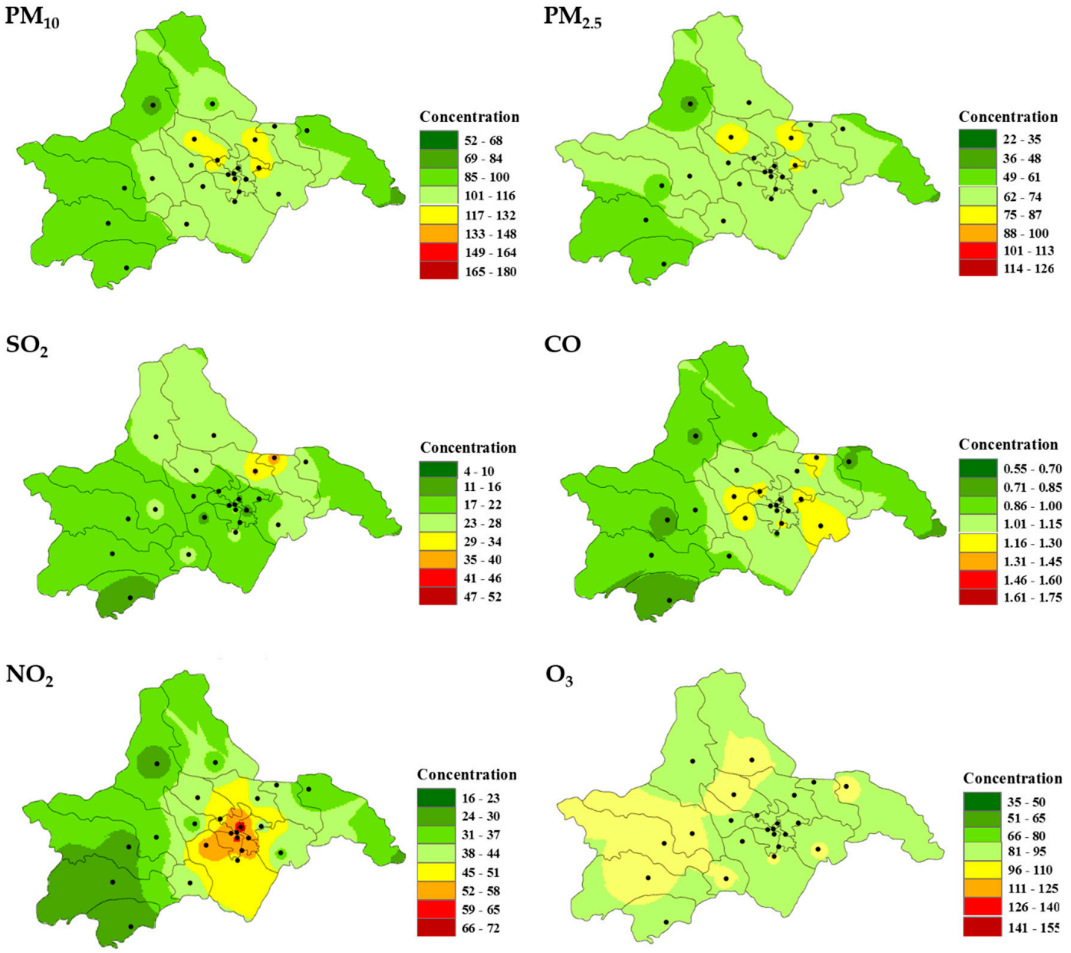

Figure 3. Spatial distributions of mean PM_{10}, $PM_{2.5}$, SO_2, CO, NO_2 and O_3 concentrations during 2014 and 2016. The units for PM_{10}, $PM_{2.5}$, SO_2, NO_2, and O_3 are μg m^{-3}, whereas that for CO is mg m^{-3}.

The average CO concentration was higher in the middle region of Chengdu, especially areas S9, S10 and S14, which are closely linked with a large population and polluting activities in the region such as a large number of cars and industrial enterprises. The CO concentration exhibited a moderate decrease from 2014 to 2016, unlike the marked decrease in SO_2. The mean concentrations of CO decreased from 1.143 ± 0.211 mg/m^3 to 1.021 ± 0.136 mg/m^3. The predominant anthropogenic CO originated from the combustion of biomass and fossil fuels. The energy consumption structure in Chengdu has changed significantly in recent years, which could decrease the emission of CO [41]. For example, straw recycling is now conducted instead of open burning of fuels, and residents are prohibited from using coal and encouraged to replace it with natural gas, which has led to less CO being released (http://www.cdstats.chengdu.gov.cn/).

In general, the spatial distribution of NO_2 and O_3 concentration is opposite: areas with high (low) NO_2 concentrations tend to have lower (higher) O_3 concentrations (especially in central Chengdu). The spatial distribution characteristics of O_3 and NO_2 concentrations reflect the complex non-linear relationship in O_3 formation with NO_2. The mean concentrations of NO_2 and O_3 displayed opposite inter-annual variation to other gaseous pollutants, which increased from 41 ± 14 μg/m^3 and 83 ± 10 μg/m^3 in 2014 to 43 ± 12 μg/m^3 and 98 ± 11 μg/m^3 in 2016, respectively (Figure 3, Table S1). It is well documented that industrial activities and vehicle emissions are major contributors to NO_2 in

China [42]. In Chengdu, the number of automobiles linearly increased from 2.13 million in 2013 to 4 million in 2016, with a striking growth rate of 29.3% each year (http://www.cdstats.chengdu.gov.cn/). However, the NO_2 emissions per automobile decreased slightly following the upgrade of oil product quality standards [43]. In addition, many factories were obliged to employ low-NO_2 burner technologies and import denitrification facilities after the implementation of emission standards for coal-fired power plants. Recently, an updated standard (GB13223-2011) with more stringent emission limits (100 mg/Nm^3) has been proposed, demanding that all newly built plants and most of the in-use plants must install advanced SCR or SNCR devices to reduce NO_2 emissions [44]. Thus, the contribution of vehicle emissions was probably counteracted by controlling the NO_2 emissions, thereby leading to the non-significant increase in NO_2 concentration in ambient air.

O_3 is a secondary pollutant that is generally formed in the atmosphere through photochemical pathways of NOx and volatile organic compounds (VOCs). Recently, a marked increase in overall O_3 was observed, although the NO_2 concentration remained steady. This may be attributed to the considerable increase in VOCs emission in some industrialized areas, leading to an elevation in O_3 through reaction with NOx. For example, a petrochemical project with an annual output of 0.8 million metric tons of ethylene started operation in 2014; it is located in the territory of Pengzhou City (S17) in the north of Chengdu. The prevailing wind direction in most parts of Chengdu is NE and NNE. In addition, vehicle emission is also the main source of non-methane volatile organic compounds (NMVOCs). The study found that the relative contribution of NMVOCs emitted by vehicles reached 52–69% in Beijing [45]. A significant increase in NMVOCs, which can react with NO_x, leads to an increase in O_3 concentration, although the NO_2 concentration increased slightly in recent years. In addition, the increase in NO_2 concentration in the center of Chengdu (e.g., S1, S3, S5–S8 and S14) is probably due to the titration reaction of NO and O_3, at the same time resulting in a decrease in O_3 concentration in this region.

3.2.2. Seasonal Variation in Air Pollutants

To evaluate air pollution events in Chengdu, spatial patterns of six pollutants (PM_{10}, $PM_{2.5}$, SO_2, NO_2, O_3 and CO) in four seasons are shown in Figure 4 (the seasonal averages at all sites are shown in Table S3). In addition, monthly trends at the sites are shown in Figure 5. All sites showed considerable seasonal variation in PM_{10}, $PM_{2.5}$, NO_2, O_3 and CO levels, with the exception of SO_2.

The average $PM_{2.5}$ concentration exhibited great temporal variability, with the highest value in winter and the lowest value in summer; the spring value was slightly higher than that in autumn. The elevated $PM_{2.5}$ concentrations in winter were the result of stationary sources (e.g., industries) and mobile sources (e.g., vehicles), unlike the northern cities in China, where the elevated $PM_{2.5}$ concentrations in winter were the result of coal combustion and biomass burning for residential heating (as most southern cities such as Chengdu did not have central heating in winter) [46]. In addition, less precipitation, lower temperature and boundary layer height, and weaker winds in winter may further exacerbate ambient pollution [47,48]. Secondary sulfate and nitrate tended to accumulate in winter due to lower boundary layer height and precipitation, and higher humidity [31,48]. The $PM_{2.5}$ concentrations in winter peaked in regions S14 and S2, and industrial sources could play significant roles in $PM_{2.5}$ accumulation due to the presence of more factories [49]. In addition, biomass burning (e.g., open burning of straw) probably led to elevated $PM_{2.5}$ in spring and autumn [32].

The PM_{10} concentration followed the order: winter (153 ± 18 μg/m^3) > spring (114 ± 11 μg/m^3) > autumn (87 ± 14 μg/m^3) > summer (73 ± 11 μg/m^3), similar to the seasonal variation in $PM_{2.5}$. Apart from the combined impacts of industrial sources and unfavorable weather conditions on air pollution dilution and dispersion in winter, this will also be severely affected in spring by dust from the north. Researchers studied the input of dust in Chengdu in March 2013 and found that the concentrations of PM_{10} and $PM_{2.5}$ rapidly increased and the ratio of $PM_{2.5}$ to PM_{10} was only 0.28 [46,50].

CO and NO_2 also exhibited similar seasonal variations, with the highest concentrations in winter and the lowest in summer. These seasonal variations reflect the effects of meteorological conditions and

emissions. For example, stagnant meteorological conditions characterized by slow winds and shallow mixing layers occur more frequently in winter, trapping the pollutants near the surface and leading to high concentrations [51]. In addition, fossil fuel combustion sources such as residential coal and biomass combustion for heating also contributed to the formation of high pollution in winter [52,53]. In contrast, solar radiation, strong turbulent eddies, increased photochemical activity and precipitation scavenging diluted the pollutants released at the surface and caused lower CO and NO_2 concentrations in summer [54]. CO originated from biomass burning, including open crop straw burning, which often occurs in autumn and is the main reason for the higher CO concentration in autumn than in spring. The NO_2 concentrations in S1 were the highest among the sites every season. Mean NO_2 concentrations were 65 μg/m^3 (spring), 56 μg/m^3 (summer), 60 μg/m^3 (autumn), and 71 μg/m^3 (winter), which were 56.1%, 67.4%, 55.3%, and 47.8% higher than mean values in corresponding seasons among the remaining sites in Chengdu.

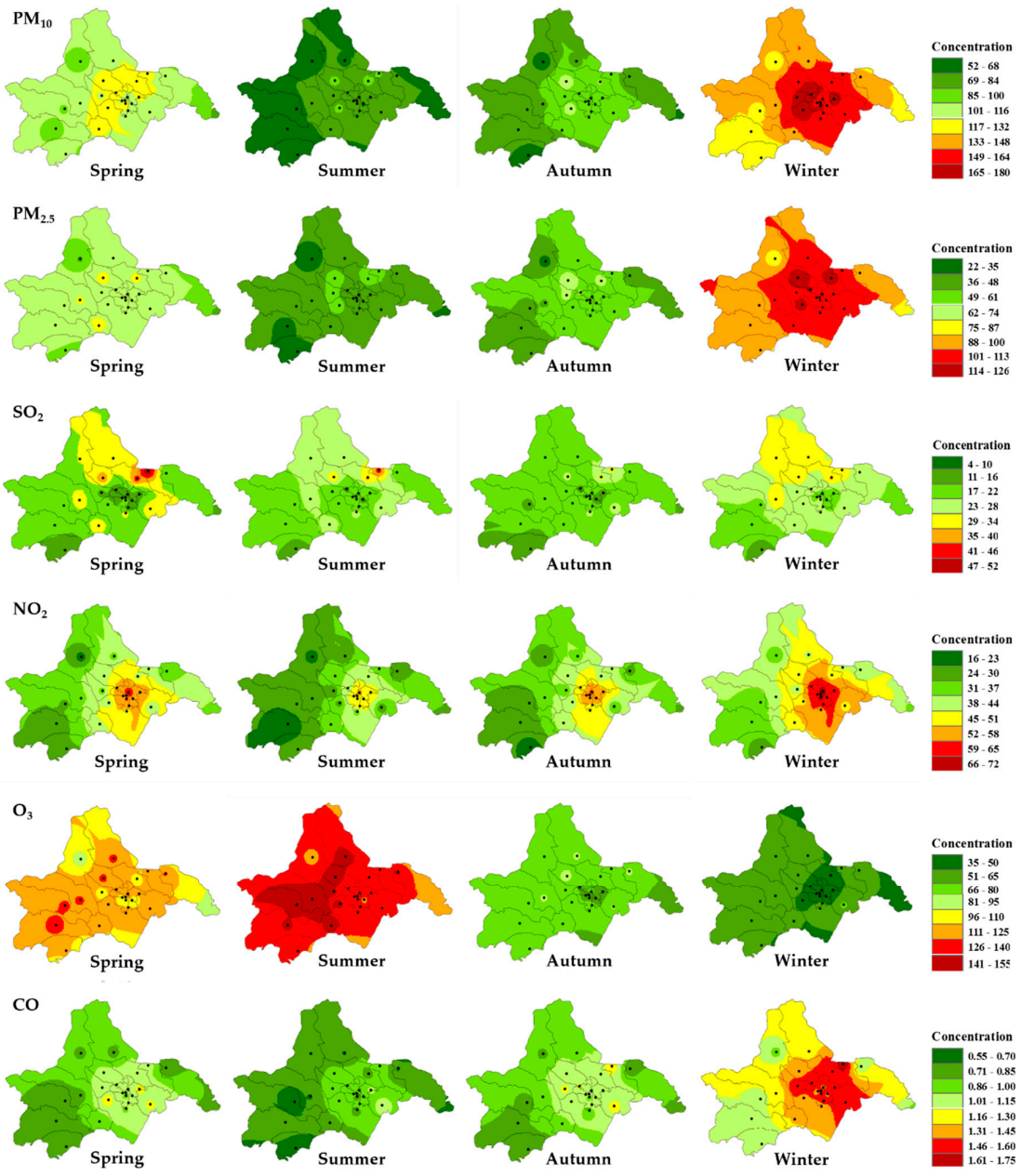

Figure 4. Seasonal variation in air pollutants in Chengdu during 2014–2016. The units for PM_{10}, $PM_{2.5}$, SO_2, NO_2 and O_3 are μg m^{-3}, whereas that for CO is mg m^{-3}.

However, the concentration of O_3 displayed the opposite seasonal variation, with the highest value in summer (136 ± 9 μg/m^3) and the lowest value (53 ± 9 μg/m^3) in winter. Weak sunlight in winter could inhibit the formation of O_3 as the formation rate of O_3 depends on the intensity of solar radiation [55]. In contrast, high temperature and strong solar radiation tended to generate large numbers of OH radicals, resulting in the formation of O_3 through the reaction of VOCs and OH radicals [56]. However, no marked increase in O_3 was observed in winter, suggesting slow photochemical activity in winter [57].

SO_2 concentrations showed weak seasonal variability: winter (25 ± 6 μg/m^3) > spring (22 ± 7 μg/m^3) > summer (20 ± 8 μg/m^3) > autumn (19 ± 5 μg/m^3); the summer and autumn concentrations were approximately the same. The main source of gas-phase SO_2 was from the combustion of all sulfur-containing fuels (oil, coal, and diesel). Furthermore, a smaller variability in monthly average SO_2 was noted during autumn, which was indicative of lower near-surface anthropogenic sources and boundary layer height evolution.

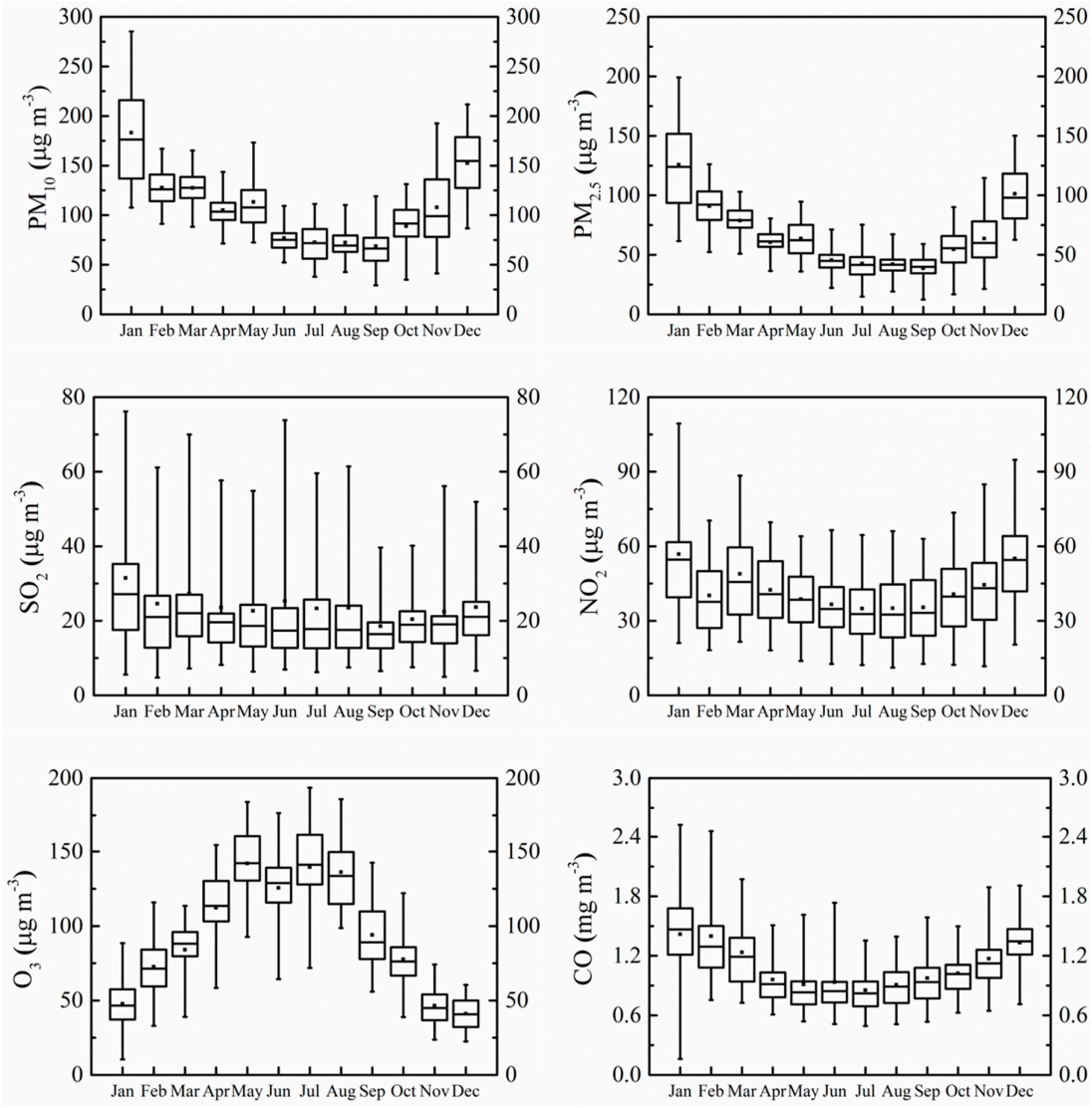

Figure 5. Monthly variations in PM_{10}, $PM_{2.5}$, SO_2, NO_2, O_3, and CO. The horizontal solid lines indicate the median and the filled squares indicate the mean, and range where the top and bottom of the box indicate the 75th and 25th percentiles, respectively. The top and bottom whiskers indicated the maximum and minimum values, respectively.

3.3. Effects of Meteorological Factors on the Air Pollutants

Unlike long-term changes or annual and diurnal variations, daily fluctuations in air pollutant concentrations may primarily depend on synoptic-scale weather conditions, especially on polluted days, assuming that the pollutant emissions remained almost constant each day for most of the study period. To quantitatively investigate the relationships between daily fluctuations in air pollutant concentrations and meteorological factors in exceedance days (according to the CAAQS standard), the correlation coefficients between the PM_{10}, $PM_{2.5}$, and O_3 concentrations and meteorological factors were calculated and are shown in Table 3 (NO_2 samples were too small to be statistically significant). The single-sample K–S test demonstrated that all sample data obeyed a normal distribution at the significance level of 0.01.

Of these meteorological factors, T, P, and RH were the most closely correlated with PM_{10} and $PM_{2.5}$, and only T was significantly positively correlated with O_3. Significant positive or negative correlations are a reflection of the physical response mechanisms. For example, the significant negative correlations with T ($p < 0.01$) and positive correlations with P ($p < 0.05$) suggest that the PM concentrations increase with decreasing temperature and increasing surface pressure because particles often accumulated in the atmosphere and were not dispersed rapidly when the temperature decreased and atmospheric pressure increased, and this was one of the causes of hazy weather. For RH ($p < 0.05$), the significant positive correlations with PM_{10} and $PM_{2.5}$ were associated with windless, cloudy, and weak sunshine days, which encouraged the accumulation and chemical reaction of pollutants [58]. We also observed that there were significant negative correlations between S, Ws, R, and $PM_{2.5}$, but no close correlation with PM_{10}. This suggests that the reliance of $PM_{2.5}$ on meteorological factors such as sunshine duration, wind speed, and precipitation was more significant than that for PM_{10} during the period where the standard was exceeded, which may be related to the different physical characteristics, sources, and composition of the two pollutants. For example, secondary aerosols are important sources of $PM_{2.5}$ in Chengdu, which is more sensitive to meteorological factors. In contrast, the O_3 concentration was only significantly positively correlated with T ($p < 0.05$), as higher temperatures can promote photochemical reactions and generate abundant O_3 [59,60]. Moreover, the correlations between O_3 and S and RH were not significant, which suggests that the links between O_3, sunshine duration, and relative humidity on a daily time scale were weak on exceedance days. This may be due to the limited fluctuation of sunshine duration and relative humidity in the short-term steady state, leading to a decrease in this correlation. In general, long sunshine duration and low relative humidity usually play an important role and have a direct influence on chemical kinetic rates and the mechanistic pathways of O_3 production.

Table 3. Analysis of the relationships between air pollutants and meteorological factors based on Pearson's correlation coefficients. The correlations are expressed as Pearson's correlation coefficient, where "*" and "**" denote significant correlations at $p < 0.05$ and $p < 0.01$ (two-tailed), respectively. AWs, MWs, S, T, RH, R, and P denote the daily average wind speed, daily maximum wind speed, sunshine duration, daily mean temperature, relative humidity, daily precipitation amount, and surface pressure, respectively.

Items		AWs	MWs	S	T	RH	R	P
		(m/s)	(m/s)	(h)	(°C)	(%)	(mm)	(hPa)
PM_{10}	r	−0.096	−0.029	−0.104	−0.248 **	0.145 *	−0.120	0.175 *
	p	0.183	0.684	0.149	0.000	0.043	0.093	0.014
$PM_{2.5}$	r	−0.119 *	−0.131 *	−0.176 **	−0.324 **	0.136 *	−0.111 *	0.206 **
	p	0.025	0.013	0.001	0.000	0.010	0.036	0.000
O_3	r	−0.029	−0.099	0.161	0.197 *	−0.174	−0.117	−0.156
	p	0.765	0.308	0.095	0.040	0.224	0.224	0.105

4. Conclusions

Based on the daily air pollutant concentrations from 23 air quality monitoring stations and meteorological datasets during 2014–2016, the air pollution characteristics and influencing factors during 2014–2016 in the inland basin city Chengdu were analyzed in this study. The main conclusions are as follows:

(1) Heavy air pollution was induced mainly by high PM or ozone concentrations in Chengdu. The annual mean concentrations of $PM_{2.5}$ and PM_{10} exceeded the standards of CAAQS and WHO at all of the stations.

(2) Air pollution was regional in Chengdu, and daily mean CO, NO_2, $PM_{2.5}$, and PM_{10} inside the middle area (e.g., S1–S8) were much higher than other regions, while daily maximum 8-h average surface O_3 concentrations and SO_2 were lower inside the middle area. Furthermore, the PM_{10}, $PM_{2.5}$, SO_2, and CO concentrations decreased from 2014 to 2016; the NO_2 level basically stable, whereas the O_3 level increased markedly during this period.

(3) $PM_{2.5}$, PM_{10}, SO_2, CO, and NO_2 displayed the highest levels in winter and the lowest level in summer (SO_2 lowest in autumn), indicating the combined impact of industrial sources and unfavorable weather conditions on air pollution dilution and dispersion. However, the O_3 concentration peaked in summer, which was associated with the strong solar radiation.

(4) Meteorological conditions are important factors that affect the concentrations of air pollutants in excessive standard days. Haze pollution can be formed easily under the weather conditions of static wind, low temperature, high relative humidity, and high surface pressure inside Chengdu. In contrast, severe ozone pollution is often associated with high temperature.

(5) The results indicate that air pollution in Chengdu is caused by multiple pollutants, and the air pollution shows great divergence in different regions and different seasons. Region-oriented air pollution management plans are suggested. This study also calls for future studies to investigate the associations between air quality and meteorological conditions, emissions in different regions, transport and transformation of pollutants in both intra- and inter-regional contexts, to further improve the understanding of the physical and chemical processes that affect air quality in Chengdu. In addition, we strongly recommend a revision and update of CAAQS standards for SO_2, as the difference between the CAAQS and WHO is too large, and the assessment results are also quite different.

Supplementary Materials: The following are available online at www.mdpi.com/2073-4433/9/2/74/s1.

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