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Abstract: Sichuan Basin is one of the most densely populated areas in China and the world. Human activities have great impact on the air quality. In order to understand the characteristics of overall air pollutants in Sichuan Basin in recent years, we analyzed the concentrations of six air pollutants monitored in 22 cities during the period from January 2015 to December 2020. During the study period, the annual average concentrations of CO, NO₂, SO₂, PM_{2.5} and PM₁₀ all showed a clear downward trend, while the ozone concentration was slowly increasing. The spatial patterns of CO and SO₂ were similar. High-concentration areas were mainly located in the western plateau of Sichuan Basin, while the concentrations of NO₂ and particulate matter were more prominent in the urban agglomerations inside the basin. During the study period, changes of the monthly average concentrations for pollutants (except for O₃) conformed to the U-shaped pattern, with the highest in winter and the lowest in summer. In the southern cities of the basin, secondary sources had a higher contribution to the generation of fine particulate matter, while in large cities inside the basin, such as Chengdu and Chongqing, air pollution had a strong correlation with automobile exhaust emissions. The heavy pollution incidents observed in the winter of 2017 were mainly caused by the surrounding plateau terrain with typical stagnant weather conditions. This finding was also supported by the backward trajectory analysis, which showed that the air masses arrived in Chengdu were mainly from the western plateau area of the basin. The results of this study will provide a basis for the government to take measures to improve the air quality in Sichuan Basin.

Keywords: air pollution; spatio-temporal variations; Sichuan Basin; back-trajectory

1. Introduction

In the past 20 years, China has experienced severe air pollution due to rapid economic development and increasing urbanization [1]. Studies showed that exposure to ambient air pollution has been associated with increased risks of mortality and morbidity worldwide [2,3]. According the Global Burden of Disease (GBD) project, air pollution was responsible for 1.6 million deaths in China and 4.2 million deaths worldwide in 2015 [4]. The continuous and serious air pollution has caused an immense burden for China's medical and economic [5]. In order to cope with serious air quality problems, China has taken a series of measures in recent years [6,7].

In 2005 and 2011, China implemented the installation of desulphurization and selective catalytic reduction (SCR) systems for coal-fired power plants [8]. At the same time, the strategies of upgrading vehicle fuel and prohibiting polluting old vehicles were introduced at the city level [9]. The Ministry of Environmental Protection of China issued the revised "Ambient Air Quality Standards" (CAAQS, GB3095-2012) in February 2012, adding PM_{2.5} and O₃ to CAAQS for the first time [10]. In 10 September 2013, the Chinese government promulgated the Air Pollution Prevention and Control Action Plan. The plan aimed to reduce the number of severely polluted days drastically and improve the national air quality significantly through long-term efforts [11]. Despite these efforts, there were still many cities that have not yet reached the current CAAQS [12]. According to the "2020



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Copyright: © 2021 by the authors. Licensee MDPI, Basel, Switzerland. This article is an open access article distributed under the terms and conditions of the Creative Commons Attribution (CC BY) license (https:// creativecommons.org/licenses/by/ 4.0/). Reports on the State of Environment of China", there were still 135 cities whose ambient air quality exceeded the standard, accounting for 40.1% of the total number of cities. In the days exceeding the standard, the proportions of PM_{2.5}, O₃, and PM₁₀ as the primary pollutants were 51%, 37.1%, and 11.7%, respectively.

Previous studies showed that Beijing-Tianjin-Hebei area (BTH), Yangtze River Delta (YRD), Pearl River Delta (PRD) and Sichuan Basin were the four main regions with severe air pollution in China [13,14]. In Beijing, YRD and PRD, some scholars have carried out a lot of research to understand the basic characteristics, chemical mechanisms, main components and transmission sources of air pollution [15–19]. Since 2000, the air quality in Sichuan Basin has further deteriorated due to increased anthropogenic emissions. However, only a few studies have focused on Sichuan Basin [20,21]. And in the past, related studies on Sichuan Basin were mainly concentrated in the two megacities of Chongqing and Chengdu, and there were few studies on the overall air quality for the whole of large-scale valley terrain [22,23]. The characteristics and source of air pollution for Sichuan Basin in recent years are still unclear [24]. In this study, we analyzed air quality data collected from Sichuan Basin for six years (January 2015 to December 2020) to fill this gap. The main goal is to investigate (1) the temporal and spatial characteristics of the overall air pollution in Sichuan Basin, (2) the industry contribution reflected by the ratio of different pollutants, and (3) a regional-scale air pollution episode that influenced multiple cities in the region. The knowledge gained in this study provides a scientific basis for formulating future emission control policies aimed at reducing severe PM_{2.5} pollution in this unique watershed

2. Materials and Methods

2.1. Air Quality Monitoring Sites

The air quality was monitored at 127 stations spread over 22 cities across Sichuan Basin, covering an area of over 260,000 square kilometers. Located in the central and southern part of the Asian continent, with a total population of more than 100 million, Sichuan Basin is one of the most densely populated areas in China and the world. Completely surrounded by high mountains and plateaus, it is a vast subtropical low hills and plains. The west is surrounded by the high-altitude Qinghai-Tibet Plateau, the south is the Yunnan-Guizhou Plateau, the east is Wushan, and the north is Dabashan. Due to low wind speed and high relative humidity, it was one of the four traditional areas with acidic rain and frequent haze events [25]. Figure 1 showed the locations of the 22 cities that collected the air quality data used in this study.

Considering the completeness of the data, this study collected the socio-economic data of each city in Sichuan Basin during 2018 (source: http://tjj.cq.gov.cn/zwgk_233/tjnj/2019 /zk/indexce.htm (accessed on 15 September 2021)). Table 1 listed the city's abbreviations, number of vehicles, population, and GDP (Gross Domestic Product). In 2018, Chongqing's total population was 31.01 million, the total number of vehicles was 6.31 million, and the GDP was 20363 billion yuan, ranking first among the cities. As another megacity in Sichuan Basin, Chengdu has a total population of 16.33 million, a total of 4.87 million vehicles, and a GDP of 1.5342 billion yuan, second only to Chongqing.



Figure 1. The map of Sichuan Basin and the locations of 22 city stations.

Table 1. Urban areas	, population	number of	vehicles,	and GDP o	of each city	y in the Sichuan	Basin in 2018.
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City	Population	GDP	Primary Industry	Secondary Industry	Tertiary Industry	Vehicle Numbers
	(10,000 Persons)	(Billion Yuan)	(Billion Yuan)	(Billion Yuan)	(Billion Yuan)	(10,000 Units)
Chengdu (CD)	1633	15,342.77	522.59	6516.19	8303.99	487.7169
Mianyang (MÝ)	485.7	2303.82	301.27	929.4	1073.15	49.9193
Devang (DY)	354.5	2213.87	243.31	1071.13	899.43	40.5155
Leshan (LeS)	326.7	1615.09	165.92	721.78	727.39	39.2793
Meishan (MŚ)	298.4	1256.02	186.5	554.46	515.06	37.9811
Yaan (YA)	154	646.1	85.83	303	257.27	11.1461
Ziyang (ZÝ)	251.2	1066.53	166.79	507.61	392.13	31.0002
Zigong (ZG)	292	1406.71	151.55	653.71	601.45	66.1452
Yibin (YB)	455.6	2026.37	248.57	1006.73	771.07	51.8162
Luzhou (LZ)	432.4	1694.97	190.58	882.97	621.42	18.2872
Neijiang (NJ)	369.9	1411.75	219.31	610.8	581.64	25.1536
Chongqing (CQ)	3101.79	20,363.19	1378.27	8328.79	10,656.13	631.7233
Guang'an (GA)	324.1	1250.24	173.52	575.23	501.49	35.733
Nanchong (NC)	644	2006.03	381.87	824.05	800.11	17.9184
Suining (SN)	320.2	1221.39	165.64	565.22	490.53	25.2796
Guangyuan (GY)	266.7	801.85	118.1	358.56	325.19	24.403
Dazhou (DZ)	572	1690.17	326.24	603.91	760.02	18.0632
Bazhong (BZ)	332.2	645.88	98.27	316.39	231.22	9.0715
Aba (AB)	94.4	306.67	49.55	139.53	117.59	24.2416
Ganzi (GZ)	119.6	291.2	65.47	121.78	103.95	23.3053
Liangshan (LS)	490.8	1533.19	307.61	613.13	612.45	35.1002
Panzhihua (PZĤ)	123.6	1173.52	39.74	731.13	402.65	25.5248

2.2. Air Quality Data

The concentrations of six pollutants, SO₂, NO₂, CO, O₃, PM_{2.5} and PM₁₀, were monitored hourly over 22 cities across Sichuan Basin from January 2015 to December 2020. The data were made available by the China National Environmental Monitoring Center (http://www.cnemc.cn/ (accessed on 15 September 2021)).

The instruments for air quality monitoring were deployed according to the China Environmental Protection Standard HJ 664-2013. The equipment came from Shenzhen Aosen Purification Technology Co., Ltd., China. The gaseous pollutant and PM concentrations were measured following the Specifications and Test Procedures for Ambient Air Quality Continuous Automated Monitoring System HJ 654-2013 for SO₂, NO₂, O₃ and CO, and HJ

653-2013 for $PM_{2.5}$ and PM_{10} , as stipulated in the National Environmental Protection Standards of the People's Republic of China. The air quality monitoring stations were located at least 50 m away from any notable stationary pollution sources, and the inlets for the instruments were placed at least 1 m higher than the roof of the building or wall [26]. Data quality assurance and quality control (QA/QC) were conducted following the technical guidelines on environmental monitoring quality management (HJ 630-2011) established in the National Environmental Protection Standards of the People's Republic of China. The validity of the data was checked following the national ambient air quality standards specified in the National Standards of the People's Republic of China (GB 3095-2012), as used in earlier studies [27,28]. The daily, monthly, and annual means of the data were calculated from the hourly concentrations (with ~80% of the available data to be considered as valid for calculating the mean).

2.3. Back-Trajectory Analysis

Backward trajectory analysis essentially follows a parcel of air backward in hourly time steps for a specified length of time [29]. The HYbrid-Single Particle Lagrangian Integrated Trajectory (HYSPLIT) model developed by the National Oceanic and Atmospheric Administration (NOAA) was used to identify the potential source area of air pollution in a specific city and capture the vertical movement of the air masses from the sources to the receptor inside planetary boundary layer [30].

The HYSPLIT model was used to investigate the movement of air masses during a heavy particulate pollution observed in winter 2017. In order to understand the impact of the regional transmission process, three-dimensional 48 h backward trajectories arriving at 500 m above ground level (AGL) of the receptor sites were also calculated using $1^{\circ} \times 1^{\circ}$ Global Data Assimilation System (GDAS) data from National Centers for Environmental Prediction (NCEP). Based on the Euclidean distance between the motion trajectories, the Ward layering method was used to assign the motion trajectories to different clusters according to their moving speed and direction. The hour with the highest PM concentration of coarse particles in Chengdu was selected as the start time of the trajectory, and the backward trajectory at a height of 500 m from 3 January to 6 January 2017 was calculated. The main transportation routes that caused severe air pollution in the winter of 2017 were identified by combining the trajectory with the corresponding average concentration of pollutants [31].

3. Results

3.1. Spatio-Temporal Characteristics of the Air Quality

The annual average concentrations of the pollutants in Sichuan Basin were determined by averaging the effective data from all stations. Their values are shown in Figure 2. The annual mean concentrations of CO, NO₂, SO₂, O₃, PM_{2.5} and PM₁₀ in the entire basin area ranged from 0.67–0.90 mg/m³, 24.33–30.4 μ g/m³, 8.41–17.76 μ g/m³, 80.08–91.4 μ g/m³, 31.2–46.56 μ g/m³ and 47.87–75.19 μ g/m³, respectively. During the same period, in Chengdu and Chongqing, two megacities of Sichuan Basin, the annual average concentrations of the six pollutants ranged from 0.69–1.08 (0.79–1.1) mg/m³, 33.75–49.46 (37.18–45.5) μ g/m³, 6.56–15.75 (7.49–16.17) μ g/m³, 87.86–101.56 (68.98–81.54) μ g/m³, 39.23–61.85 (32.27–54.42) μ g/m³, 61.27–103.83 (51.85–84.12) μ g/m³, respectively.

The annual average SO₂ concentration was the lowest in Bazhong, with a value of $4.71 \ \mu g/m^3$, and the highest in Panzhihua, with a value of $33.69 \ \mu g/m^3$. The lowest NO₂ concentration of $9.21 \ \mu g/m^3$ was observed in Aba Prefecture, and the highest in Chengdu, reached $43.73 \ \mu g/m^3$. The highest annual average CO concentration observed at $1.49 \ m g/m^3$ in Panzhihua, and the lowest of $0.48 \ m g/m^3$ was observed in Ganzi. The lowest annual average concentration of O₃ was observed in Ya'an during 2015, which was $53.2 \ \mu g/m^3$. The highest annual average concentration of O₃ was observed in Zigong during 2018, with a value of $105.19 \ \mu g/m^3$. The annual average concentration of PM_{2.5} and PM₁₀ in Aba Prefecture was the lowest, $15.12 \ \mu g/m^3$ and $26.38 \ \mu g/m^3$, respectively. The



highest values of 84.94 μ g/m³ and 109.6 μ g/m³ was observed in Zigong, both of which appeared in 2015.

Figure 2. The annual average concentrations of pollutants in Sichuan Basin. The filled circle represents the mean concentration whereas the error bar denotes the range of the annual concentration. The data used in each city comes from every station during 2015–2020.

As shown in Figure 3, the concentrations of five pollutants other than O_3 all showed a downward trend from 2015 to 2020 in Sichuan Basin. The concentration of NO₂ was the highest in 2017, at 30.4 µg/m³, and the lowest in 2020, at 24.33 µg/m³, with an average annual decline rate of 2.72%. The highest concentrations of CO, SO₂, PM_{2.5} and PM₁₀ all appeared in 2015 with the value of 0.9 mg/m³, 17.76 µg/m³, 46.56 µg/m³ and 75.19 µg/m³, respectively. The lowest concentrations all appeared in 2020 with the value of 0.67 mg/m³, 8.41 µg/m³, 31.2 µg/m³, 47.87 µg/m³, and the average annual decline rates were 5.14%, 10.52%, 6.59%, and 7.27%, respectively.

At present, the environmental concentration of most air pollutants in China is declining, but the concentration of secondary pollutants such as O_3 is increasing at both provincial and capital city levels [32,33]. Previous studies showed that the rising rate of O3 in China's 2 + 26 urban areas was almost 14 times that of the global O_3 [34]. The lowest ozone concentration of 80.08 µg/m³ in Sichuan Basin was observed in 2015, and reached the highest in 2018, with the value of 91.4 µg/m³. It declined slightly in the following two years, but still showed an upward trend during the overall study period. The annual growth rate was about 0.76%.



Figure 3. Annual average concentration trend of the six pollutants.

Ozone is not directly emitted by pollution sources in the environment [35]. It is a secondary pollutant generated by chemical reactions of nitrogen oxides and volatile organic compounds under strong ultraviolet light irradiation [36]. Although China has adopted strict control measures in recent years, which has made PM, NO₂, SO₂ and other atmospheric pollutants show a clear downward trend, the ozone concentration is still slowly increasing [37]. The main reason for this phenomenon is that emissions of NOx and VOCs (main precursors of O_3) remain high in China [38]. And the meteorological conditions of high temperature and low rainfall are conducive to the generation of O_3 in recent years [39]. At the same time, the global O_3 background value has been continuously increasing, which also makes a certain promoting effect on China's ozone concentration [40].

3.2. Seasonal Variations of Pollutants

Figure 4 shows the seasonal variations of the six pollutants in each city. For almost all pollutants (except O₃), the highest concentrations were observed in winter and the lowest in summer. It is speculated that the continuous adverse weather conditions in winter include smaller wind speed and rainfall, lower temperature and atmospheric boundary height, which are not conducive to the diffusion of pollutants. And compared with other seasons, the consumption of coal and biomass fuel for heating in winter is higher [41,42]. On the contrary, in summer, the wind speed and planetary boundary layer is higher, the rainfall is abundant, the rain removal effect is obvious, and the pollutant concentration is lower [43,44].

Since NO₂ is the main man-made pollutant emitted from vehicles and transportation facilities and fuel combustion, these activities are more frequent in the two megacities of Chengdu and Chongqing than in other places [45]. In 2018, the total number of motor vehicles in Chengdu and Chongqing accounted for 28.2% and 36.5% of the entire basin area respectively. Therefore, the highest NO₂ concentration was observed in these two

cities. The average concentration of NO₂ in winter was between 111.87 μ g/m³ (Chengdu) and 50.93 μ g/m³ (Aba Prefecture). Similar characteristics were observed for CO, namely the highest and lowest concentrations were observed in winter and summer, respectively. The opposite trend of ozone occurred. The highest concentration happened in spring and summer, and the lowest concentration occurred in winter. This is related to the formation mechanism of ozone. Many studies showed that under sufficient light, volatile organic compounds (VOCs) and nitrogen oxides (NOx) underwent a photochemical reaction to generate O₃ and at the same time produced secondary pollutants in the atmosphere [46–48]. High temperature, strong ultraviolet and high photochemical reaction rate were common phenomena in Sichuan Basin during spring and summer. The weaker solar radiation in winter inhibited the photochemical reaction, which was not conducive to the production of O₃. Therefore, the O₃ concentration in the Sichuan Basin had the highest trend in spring and summer.



Figure 4. Seasonal variation in concentrations of air pollutants in Sichuan Basin. The vertical error bars represent the standard deviation values. Spring (March to March), Summer (June to August), Autumn (September to November), Winter (December to February of the following year).

Regions with high SO₂ concentrations were mainly located in the plateau areas of the western Sichuan Basin, such as Panzhihua, Liangshan, Ganzi and other cities. Far more than the cities such as Chengdu and Mianyang in the basin, the winter SO₂ concentration of Panzhihua was 38.7 μ g/m³, about 3 times of the average SO₂ concentration in whole Sichuan Basin (12.8 μ g/m³). On the one hand, the SO₂ of cities in the basin such as Chengdu was mainly derived from industrial emissions. In these areas, the government took strict desulfurization measures, which greatly reduced the concentration of SO₂. On the other hand, coal combustion for household heating due to low temperature in high altitude regions, led to more SO₂ emissions, and the implementation of desulfurization measures in these areas were not yet fully completed.

Different with northern China, due to the warm temperature in Sichuan Basin (about 10 °C on average), there was no widespread coal or wood burning for household heating in winter; therefore, atmospheric processes and meteorological conditions played an

important role in the seasonal changes of particulate matter effect [49]. Almost all regions had the highest concentration of particulate matter in winter, about 1.8–2.5 times that of the other three seasons. The concentration was similar in spring and autumn, and the lowest concentration occurred in summer.

3.3. Analysis of City's Pollutant Ratio

The average PM_{2.5}/PM₁₀ ratio of all cities in Sichuan Basin was 0.61, and the monthly ratio was between 0.43 and 0.69, appeared in April and January respectively. The average ratio in Chengdu and Chongqing was 0.60 and 0.63, respectively. In 2017, a study reported the average ratio of 0.58 for 31 provincial capital cities, and Zhang (2015) reported an average ratio of 0.56 for 190 cities in China [7,50]. However, in Beijing (0.80), Shanghai (0.70) and Guangzhou (0.72), the ratio was much higher than that observed in this study [51]. These findings indicated that, compared to developed cities in China, the air quality in Sichuan Basin was more affected by coarse particles. Figure 5 showed the monthly average ratios of different cities in Sichuan Basin during the study period. The lowest average ratio was found to be 0.43 (in Guangyuan), while the highest average ratio (0.69) was observed in Zigong and Luzhou.



Figure 5. The monthly average PM_{2.5}/PM₁₀ ratio of each city.

In winter, all cities had the highest $PM_{2.5}/PM_{10}$ ratio, while in spring and summer, the ratio decreased rapidly. This was due to the high emissions of coarse particulate matter from sand and soil during the spring when it is very dry, windy, and dusty in Sichuan Basin [52]. Dust emitted from desert areas in Xinjiang (such as Taklimakan) may be transported towards the Qinghai-Tibet Plateau in the northwest of Sichuan Basin, thereby affecting the atmosphere and ecosystems of the basin area.

 SO_2 can be used to normalize $PM_{2.5}$ to exclude the effects of coal combustion and meteorological conditions. It can be seen from Figure 6 that during the study period, among the cities in Sichuan Basin, the city with the highest $PM_{2.5}/SO_2$ was Bazhong (6.39), followed by Deyang (5.19) and Nanchong (5.18), which reflected the contribution of non-industrial source to $PM_{2.5}$. The average ratio in Sichuan Basin is 3.45, which was close to

the national average (2.92) in the previous study [53]. It is worth noting that in Panzhihua, Aba, Liangshan and other areas, the value of $PM_{2.5}/SO_2$ has always remained at a low level throughout the year, which may be because the industry in Sichuan Basin is mainly concentrated in the western region, and industrial sources contribute more to fine particles.



Figure 6. The monthly average PM_{2.5}/SO₂ ratio of each city.

 $PM_{2.5}/SO_2$ also exhibited a U-shaped mode in most cities, which reflected that nonindustrial sources such as power and residential contributed more to $PM_{2.5}$ in winter. Multi-resolution Emission Inventory for China (MEIC) was often used to estimate emissions from various sectors in China [54]. In order to determine the relationship between this dynamic change and the emission trends of key sectors involved in the air pollution process, we collected $PM_{2.5}$ emission information from key sectors in the 2017 MEIC in-ventory of Sichuan Basin (Figure A1). Among them, non-industrial source emissions showed a similar U-shaped trend, which was consistent with the previous conclusions.

CO is an indicator of the primary combustion source. The secondary formation of fine particles in the basin can be studied by calculating the ratio of $PM_{2.5}$ to CO [55]. From 2015 to 2020, the value of $PM_{2.5}/CO$ was higher in the southern areas such as Luzhou and Zigong, and the lowest in the western plateau areas such as Panzhihua (Figure 7). This indicated that the secondary sources in the southern cities of the basin had a higher contribution to the generation of fine particles.

Previous research reported that the sulfur dioxide emissions were much lower than the emissions of nitrogen oxides for motor vehicles in China, and the ratio of $[SO_2]/[NO_2]$ in motor vehicles was usually between 0.0084 and 0.042. Both NOx and SO₂ were emitted from stationary sources, but the emissions of SO₂ was relatively more. The ratio of $[SO_2]/[NO_2]$ in fixed sources was usually between 1.25 and 5 [56]. Therefore, the SO₂/NO₂ ratio was often used as an indicator of air pollution caused by stationary sources and mobile sources [57]. Figure 8 showed the monthly average ratio of SO₂/NO₂ in each city. In study area, Liangshan and Panzhihua had the highest SO₂/NO₂ ratios, indicating that the air pollution in these western plateau cities mainly came from local industrial sources and coal combustion. Bazhong was the lowest (0.21), followed by Chengdu (0.24) and Chongqing (0.26). These results confirmed that there was a strong correlation between air pollution and automobile exhaust emissions in Chengdu and Chongqing.



Figure 7. The monthly average PM_{2.5}/CO ratio of each city.



Figure 8. The monthly average SO₂/NO₂ ratio of each city.

From 2015 to 2020, the ratio of $PM_{2.5}/SO_2$ in Sichuan Basin had shown a continuous upward trend, and the ratio of SO_2/NOx had shown a continuous downward trend (Figure A2). It showed that the contribution of industrial sources to fine particulate matter con-tinued to decline. This was related to the pollutant emission reduction measures that the government had introduced. $PM_{2.5}/CO$ also showed a downward trend, reflecting the de-cline in the contribution of secondary sources to fine particulate matter, which was related to the decrease in the concentration of SO_2 and NOx in the regional atmosphere. In 2015, the executive meeting of the State Council of China decided to implement ultra-low emis-sion and energy-saving retrofits for coal-fired power plants before 2020. Sichuan

Province had successively formulated the implementation rules for the Air Pollution Prevention and Control Action Plan from 2014 to 2017, and proposed a series of measures to improve the atmosphere environmental quality, including the elimination of coal-fired boilers below 10 tons per hour and the prohibition of new coal-fired boilers below 20 tons per hour. At the same time, we also noticed that during the study period, the ratio of $PM_{2.5}/PM_{10}$ showed an overall upward trend. This reflected the effectiveness of current dust removal measures to a certain extent, because the existing dust removal measures had far greater removal effects on coarse particles than fine particles.

3.4. Characterization of an Air Pollution Episode in Winter 2017

During this research period, the particulate pollution incident that caused a wide range of impacts was identified in the winter of 2017. Figure 9 showed the hourly average PM_{10} concentration of four cities affected by air pollution incidents (3–6 January 2017), during which the hourly average PM_{10} concentration of all cities exceeded 120 µg/m³. The average daily concentrations from 3–6 January in Chengdu, Deyang, Ya'an and Meishan were 366 µg/m³, 245 µg/m³, 232 µg/m³ and 225 µg/m³, respectively. The average daily PM_{10} concentration in these cities was about 10–18 times higher than the WHO guidelines. During the period of heavy pollution, the average ratio of $PM_{2.5}/PM_{10}$ increased over Chengdu, Deyang, Ya'an and Meishan to 0.65, 0.68, 0.69 and 0.74, respectively. The ratio of the four cities is greater than 0.65, much higher than the annual average value of 2017 (0.59), indicating the dominance of fine particulate matter during the event.



Figure 9. Air transport clustering trajectory in Chengdu during heavy pollution period, and the color of red and orange represent two different trajectories. The right picture shows the hourly average PM_{10} concentration of the four cities during the event. The shaded part represents the arrival time of the peak concentration.

After clustering the downloaded backward trajectories, it was found that they mainly originated over the plateau area of the southwestern part in the basin. The air mass reached Ya'an first from the southwest. The concentration of particulate matter in Ya'an reached a peak with the value of 316 μ g/m³ at 20 o'clock on 4 January, and then the air mass continued to move northeastward. When the air mass moved to Chengdu, the concentration of particulate matter increased. Moreover, the highest concentration was

observed in the Chengdu area, and the highest concentration may occurred over Chengdu and Deyang. Chengdu and Meishan peaked at around 15:00 on 5 January, with PM_{10} concentrations of 478 μ g/m³ and 288 μ g/m³, respectively. Deyang reached the maximum concentration of 324 μ g/m³ during the pollution period at around 20 o'clock on 5 January.

It is worth noting that the backward trajectory changed the direction in DY and MY before arriving in CD. The further enrichment of particulate matter concentration from DY to MY and CD may be affected by climatic conditions. The adverse meteorological conditions in heavy pollution days, including high pressure, weak wind (0.7 m/s in average) and low temperature (10.5 °C in average), make the pollution track not easy to spread and can only flow inside the basin.

Southwest region is the industrial concentration area of Sichuan Basin, with developed secondary industry. In these cities of Southwest region, PZH is one of the four major iron ore areas in China. In 2018, the economic proportion of the secondary industry in PZH was 62.3%, the highest among all cities in Sichuan Basin. Previous studies have shown that there is a strong positive correlation between the secondary industry and PM_{2.5} concentration [58]. Dense industrial sources in southwest region and adverse meteorological conditions may be the main causes of heavy pollution events in the selected cities.

3.5. Comparison of Air Quality with Standards and Guidelines

In this section, we compared the mean concentration of the pollutants with the available national and WHO guidelines to determine the impacts of current air quality on human health in Sichuan Basin. China revised the National Ambient Air Quality Standard in February 2012. The WHO standards were more stringent than China. Table 2 compared the annual average concentrations of the four pollutants in the basin with different standards, such as the United States Environmental Protection Agency (USEPA), European Union (EU), Australia and Indian standards. The situation regarding pollutants in Sichuan Basin was severe. The WHO guideline for $PM_{2.5}(PM_{10})$ was exceeded by a factor of approximately 4 (3.8), indicating that the health of the residents will be affected. And the concentrations of $PM_{2.5}$ and PM_{10} are approximately 2 and 1.8 times higher than the national Grade-I standards, respectively.

Spacias	China		WILLO	LICEDA	EU	A	T. 1.	Sichuan Bacin (This Study)	
Species	Grade-I	Grade-II	WHO	USEPA	EU	Australia	India	idia Sicilian Dasin (This Study)	
PM _{2.5}	15	35	10	15	25	8	40	39.75	
PM_{10}	40	70	20	-	40	25	60	75.19	
SO ₂	20	60	-	-	-	20 *	50	17.76	
NO ₂	40	40	40	53 *	40	30 *	40	28.17	

Table 2. Comparison of the annual average concentration of the four pollutants with the available standards.

Values are in μ g m⁻³, * Values in ppb (parts per billion).

In Figure 10, the annual average concentration was compared with WHO standards and national Grade-I and II standards. During the study period, the average annual SO_2 concentration of all cities in Sichuan Basin reached the national Grade-II standard. All cities except Panzhihua reached the national Grade-I standard in 2020. The high concentration of SO_2 in Panzhihua may be due to the burning of coal and biomass and the work of power plants. Moreover, the annual average concentration in Panzhihua declined rapidly since 2018, and it was only slightly higher than the national Grade-I standard in 2020.

During the study period, the annual NO₂ concentration in almost all cities was lower than the WHO and national Grade-I standard. As the two megacities in the basin, Chengdu and Chongqing have the largest anthropogenic activities and emissions, so their annual average concentration of NO₂ was the highest. In 2015–2018, the annual average level was 1.04–1.23 times higher than the WHO guidelines. However, in 2019–2020, its concentration dropped rapidly, mainly due to the strict implementation of the government's environmental protection policy.



Figure 10. Annual mean concentrations of the pollutants at each site and comparison with the WHO guideline values (yellow shading) and National Grade-I (black shading) and Grade-II (dark gray shading) standards for air quality.

In 2020, the concentration of fine particulate matter in almost all cities in the basin exceeded the WHO regulations. Among these 22 cities, only the average $PM_{2.5}$ concentration of Ganzi was within the WHO standard in 2020. The $PM_{2.5}$ concentration in Aba Prefecture in the past two years was only 0.37 μ g/m³ higher than the national Grade-I standard. In Chengdu, Deyang, Zigong and other cities, although the concentration of particulate matter has been declining in recent years, it was still higher than the national Grade-II standard, about 3.5–3.9 times higher than the WHO standard. Compared with $PM_{2.5}$, the situation of PM10 is slightly better. In 2020, the PM10 concentration of all cities reached the national Grade-II standard, and only Ganzi reached the WHO standard in 2019–2020.

4. Conclusions

This study used air quality monitoring data to present the overall air quality status of 22 cities in Sichuan Basin from January 2015 to December 2020. The annual average concentrations of CO, NO₂, SO₂, O₃, PM_{2.5} and PM₁₀ in the entire basin were 0.79 mg/m³, 28.17 μ g/m³, 13.08 μ g/m³, 84.76 μ g/m³, 39.75 μ g/m³ and 63.56 μ g/m³, respectively. Except for O₃, the annual average concentration of the other five pollutants showed a clear downward trend. CO, NO₂, SO₂, PM_{2.5} and PM₁₀ decreased by 25.7%, 13.6%, 52.6%, 32.9%, and 36.3% respectively during the study period. And O₃ was slowly increasing at an average annual rate of 0.6 μ g/m³. The spatial patterns of CO and SO₂ were similar. High-concentration areas were mainly located in the western plateau of Sichuan Basin, while the concentrations of NO₂ and particulate matter were more prominent in the urban agglomerations inside the basin.

The annual average value of $PM_{2.5}/SO_2$ has been maintained at a low level in Panzhihua (0.9), Liangshan (1.2) and other regions for many years, indicating that industrial sources in the western Sichuan Basin have made a greater contribution to fine particulate matter. Non-industrial sources such as electricity and housing contribute more to fine particulate matter in winter. $PM_{2.5}/CO$ is higher in the southern Sichuan Basin, such as Luzhou (0.077) and Zigong (0.075), indicating that secondary sources have a greater impact on the generation of fine particles. The low SO_2/NO_2 values in megacities such as Chengdu (0.24) and Chongqing (0.26) indicate that there is a strong correlation between air pollution and automobile exhaust emissions.

During the heavy pollution incident in the winter of 2017, the average daily concentrations from 3–6 January in Chengdu, Deyang, Ya'an and Meishan were 366 μ g/m³, 245 μ g/m³, 232 μ g/m³ and 225 μ g/m³, respectively, which were mainly caused by the surrounding plateau terrain under typical stagnant weather conditions. This finding is also supported by backward trajectory analysis, indicating that the air masses arriving in Chengdu are mainly from the plateau area in the western part of the basin. During the study period, the annual average concentration of PM_{2.5} (PM₁₀) exceeded the WHO guidelines by as much as 4 (3) times. This shows that PM is still the main air pollutant of concern in the region. Therefore, reducing PM should become an integral part of the strategy, policy and action plan of the air pollution management plan. This paper conducts an in-depth study on the temporal and spatial distribution characteristics of six standard air pollutants in the Sichuan Basin, hoping to provide a strong scientific basis for effective air pollution control planning in this area and similar urban agglomerations.

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Appendix A

Figure A1. $PM_{2.5}$ emissions of various departments in Sichuan Basin based on 2017 MEIC inventory statistics. The transportation department corresponds to the left axis, and the other departments correspond to the right axis.



Figure A2. Annual change in the proportion of pollutants.

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