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Research

Variation characteristics and coordinated emission reduction of air pollutants in megacity of Chengdu-Chongqing economic circle under dual carbon goal

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ABSTRACT

The air pollution process and impact mechanism of megacities located in complex terrain are particularly complex. As a typical megacity in China, Chengdu has unique topographical and meteorological conditions, and its pollution control is difficult. This study systematically elucidated the variation characteristics of six criteria air pollutants in Chengdu between 2014 and 2020. Besides, the $PM_{2.5}/PM_{10}$ and NO_2/SO_2 ratios were discussed. Furthermore, a detailed analysis of the correlation between air pollutants was carried out. Finally, the collaborative path of carbon reduction and air pollution control is discussed. The results indicated that SO_2, $PM_{2.5}$, PM_{10} and CO were significantly decreased by 62.9%, 50.8%, 45.5%, and 36.7%, respectively. $PM_{2.5}$ and O_3 compliance rates are very low, and O_3 increases with fluctuations. SO_2, NO_2, CO, PM showed a "U-shaped" seasonal variation, and there was a "seesaw" phenomenon between O_3 and $PM_{2.5}$. The continuous changing trends also found in the ratios of $PM_{2.5}/PM_{10}$ and NO_2/SO_2. The results highlight the importance of coordinated reduction of carbon emissions and pollutants in Chengdu. This research can improve the prediction accuracy of air pollution in complex terrain areas under global warming, and improve the understanding of the formation mechanism of air pollution in special terrains around the world.

Keywords: Atmospheric pollutant, Cheng-Yu dual-city economic circle, Carbon mitigation, Dual carbon strategy, Multi-pollutant jointly control, Variation characteristics

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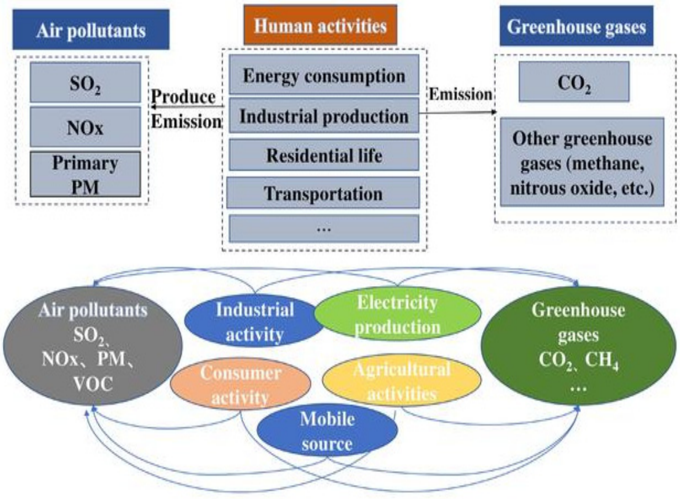
Graphical Abstract



1. Introduction

Currently, facing the enormous risks of COVID-19 and climate change, the world needs a sustainable recovery to jointly address both challenges [1, 2]. Air pollution not only accelerates the global and regional climate crisis, but also poses a huge socioeconomic threat and has been recognized as a serious public health risk [3-7]. Recent population research have revealed that $PM_{2.5}$ exposure is related to reproductive and neurological diseases, while O_3 exposure is associated with cardiopulmonary disease and lung cancer [8, 9]. Previous studies have shown that about 7 million premature deaths per year are due to air pollution [10, 11]. Previous studies also indicate that about 98% of cities in developing countries fail to fulfill the air quality guidelines of World Health Organization (WHO) [12, 13]. In addition, there is a complex connection between climate change and pollutants, and climate change affects air quality through physical, chemical and biological processes (Fig. S1) [14]. Tropospheric ozone can cause climate warming, and aerosols absorb and scatter shortwave and longwave radiation, affecting cloud radiation characteristics and precipitation characteristics, thus leading to climate change [15, 16]. While climate change (such as wind speed, temperature, and rainfall) alters the transport of atmospheric pollutants, chemical reaction rates of secondary pollutants, and wet removal of aerosols by precipitation [17]. In addition, climate change also affects dynamic vegetation and the corresponding Volatile Organic Compounds (VOCs) emissions.

As one of the countries with the fastest growth rate of pollutant emissions in the world, China's air pollution is characterized by many types of pollutants, high pollutant concentrations, and complex sources of pollution [18, 19]. It is estimated that about 0.5-1.6 million premature deaths every year are caused by air pollution in China [20-24]. In 2020, the number of premature deaths caused by long-term and short-term exposure to O_3 was 148,000 and 80,000, respectively, an increase of 49% and 51% compared with 2013 [25]. Air pollution in China has attracted worldwide attention. Faced with this daunting challenge, the Chinese government has taken various measures to reduce air pollution (Table S1). Compared with 2019, the national average concentrations values of PM_{10}, $PM_{2.5}$, O_3, NO_2, SO_2 and CO in 2020 dropped by 11.1%, 8.3%, 6.8%,11.1%, 9.1% and 7.1%, respectively [26, 27]. However, the problem of structural pollution in China is still prominent. Based on the evaluation of the new WHO Global Air Quality Guidelines (WHO, 2021AQGs), only 18.9% of the 337 cities in China reached the standard in 2020 [28, 29]. With the deepening of China's pollution control process, the space for pollutant emission reduction has narrowed significantly, and the difficulty of terminal treatment and emission reduction has gradually become prominent. In addition, the Chinese government has pledged to achieve carbon peaking and carbon neutrality targets by 2030 and 2060, which provides a new impetus for coordinating air pollution control and greenhouse gas reduction [1, 30].

Due to high energy consumption and explosive traffic growth, regional pollution incidents occur frequently in China [31-36]. Air pollution in complex terrain has attracted special attention all over the world [37-39]. It has also been widely noted that serious air pollution event is usually seen in industrialized and highly urbanized mountain (basin) cities, such as Beijing, Chengdu in China [40-43]. The complex basin topography and special meteorological conditions have accelerated the deterioration of air quality in the Chengdu-Chongqing region, with the regional average annual haze days reaching 68.7 days. Extensive studies show that the Chinese government faces huge challenges in improving air quality in Sichuan Basin (SCB) and should pay more attention to reducing pollutant emissions in megacities such as Chengdu, Chongqing [44-49]. Chengdu is one of the most important industrial production bases and key transportation hubs in the Cheng-Yu region. The land area accounts for about 2.9% of Sichuan Province (485,000 km^2), which is larger than that of key cities such as

Shanghai and Tianjin (Fig. S2). The urbanization rate reached 78.8%, and there were large differences in population density among districts and counties (Fig. S3). Besides, Chengdu's economy, urbanization rate, population and car ownership all showed a trend of rapid growth. The accelerated growth of the economy and population has also led to a rapid increase in urban electricity consumption (Fig. S4). The climate in this area is featured by subtropical monsoons, with abundant rainfall and high relative humidity (82%). The unique basin topography and adverse meteorological conditions, coupled with high local emissions, impedes the diffusion and migration of local pollutants, and the region is particularly polluted [41, 50-51]. Besides, the Cheng-Yu region has introduced many air pollution control measures to ameliorate air quality in Chengdu [52-60] (Table S2).

Coordinating the reduction of CO_2 and pollutant emissions is an inevitable choice for China's medium and long-term climate and environmental governance [61, 62]. If the emission reduction measures lack synergy considerations, it may lead to the contradiction between pollution control and carbon reduction. In fact, the distribution of population, industries, and transportation in China is extremely uneven, and there are distinct spatial and temporal differences in air pollution. However, most of the researches focus on the heavily polluted areas with developed economy such as Pearl River Delta (PRD), Beijing-Tianjin-Hebei (BTH) and Yangtze River Delta (YRD) [63-69]. There are relatively few studies on air pollution in the Cheng-Yu area, one of the four heavily polluted areas. In special terrain such as basins and valleys, the task of high-efficiency control of environmental pollution in megacities is even more arduous. Due to the unique terrain and meteorological factors, rapid economic development and high emission sources, it is critical to explore the characteristics and formation mechanism of air pollutions in Chengdu [10, 70-72]. What's more, most of the existing studies are mostly limited to single pollutant (PM or O_3) and short-term (1-2 years of heavy pollution) studies, or focus on measurements of short-term events or case studies. Few studies have focused on the long-term variation characteristics of multiple pollutants in SCB' megacities and the synergistic effect of "pollution reduction and carbon mitigation".

Cities are the basic unit of atmospheric environment management and low-carbon practice, and are the main battlefield for China to coordinately promote air pollution control and climate change response, strive to achieve "carbon peak" by 2030. Under the background of "One Belt, One Road", "Chengdu-Chongqing Economic Circle" and "Dual Carbon" goals, choosing Chengdu, a typical megacity in Cheng-Yu region, as the research object has theoretical prospective value. More importantly, exploring the long-term changes in the concentrations of various pollutants and their coordinated emission reductions of greenhouse gases and air pollution can improve the prediction accuracy of regional air pollution in areas with complex terrain under global warming, even broaden the understanding of formation mechanism of air pollution in the special terrain of the world. The coordinated management of greenhouse gases and air pollutants is a "win-win" measure for the improvement of population health. This research will provide a great impetus for promotes related health risk assessments epidemiological research.

Based on previous studies, this paper comprehensively investigates the air quality status in Chengdu during 2014-2020. Then the long-term annual, quarterly and monthly evolution of six standard air pollutants ($PM_{2.5}$, PM_{10}, CO, SO_2, NO_2 and O_3) was systematically elucidated through statistical analysis methods, and combined with Chinese Ambient Air Quality Standards (CAAQS) and the latest WHO, 2021AQGs standards excessive levels of pollutants were judged. In addition, the variation features of $PM_{2.5}/PM_{10}$ and NO_2/SO_2 ratios was emphatically discussed. Furthermore, a comprehensive and detailed analysis of the correlation between six air pollutants was carried out. Finally, based on the "dual-carbon" goal, the collaborative path of greenhouse gas reduction and air pollution control was discussed. Compared with existing studies, the research period of this study is longer (6 years), the research scale is more refined (different time scales and site scales), and the pollutants studied are more comprehensive (six criteria pollutants). Therefore, it can provide a reference for local governments to formulate more precise and effective air pollution control measures.

2. Methodology

2.1. Study Area

The Cheng-Yu region (also known as the SCB) a lowland region in southwest China (25°~35° N, 95°~110°E), and is the intersection of the "Belt and Road" and the Yangtze River Economic Belt [45, 47, 73]. This region is topographically isolated and is located in one of the most topographically complex areas in the world [44-49]. It is located at the altitude between 250 m and 750 m, and it is completely encircled by topography of mountains and plateaus (Fig. S5) [74]. Furthermore, the unique climatic features of this region are extremely low wind speeds (0.9-1.4 m/s), a high frequency of atmospheric inversions and high relative humidity (79-84%) all year round [10, 75-77]. The closed environment makes the stability of the atmospheric stratification in the basin boundary layer higher than that in other areas at the same latitude. In addition, the frequency of static winds in the basin is high, which blocks the diffusion of air pollutants, resulting in the continuous accumulation of air pollutants in the basin and remaining high concentrations. Chengdu and Chongqing are two biggest core megacity in Cheng-Yu region, located in its plat west and mountainous east, respectively and become the two most concerned cities in this region [40, 78-79].

Chengdu is located in southwest China (30.08°~31.43°N, 102.9°~104.88°E), in the hinterland of the Chengdu Plain, with a total area of 14,335 km^2 and an average altitude of 500 meters [80-82]. It is the capital city of Sichuan province as well as one of the largest cities in Western China and is encircled by the Longquan Mountains to the east and the Qionglai Mountains to the west [53, 80-84]. The city has a total of 20 county-level administrative districts, including the central urban area and the main urban area [80-82]. In addition, the terrain of this area is high in the west and low in the east, with a height difference of 4,966 meters, and the terrain is relatively closed [41, 56, 58, 75]. Due to the huge vertical height difference, unique landform types of plains

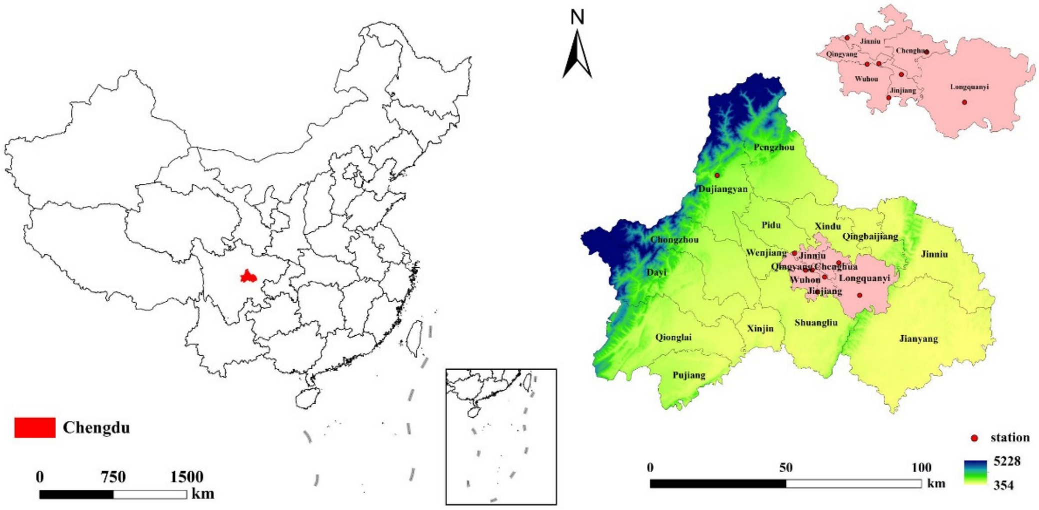


Fig. 1. Location of the eight automatic ambient air monitoring stations

(40.1%), hills (27.6%) and mountains (32.3%) are formed in this area. More importantly, the annual average temperature and sunshine from 2010 to 2020 were 15.2°C-16.6°C, 1690 h respectively (Fig. S6). The wind speed is about 1.2 m/s, the static wind frequency is as high as 45% to 50%, and the temperature inversion occurs frequently [85, 86]. Stationary and stable winds will inhibit the diffusion of atmospheric pollutants in the area, accelerate the accumulation of pollutants, and lead to an increase in pollutant concentrations.

2.2. Data Collection

The daily average concentrations of six air pollutants were derived from eight state-controlled monitoring stations in Chengdu (Junpingjie, Liangjiaxiang, QinjuanLianghe, Caotangjie, Shilidian, Sanwayao, Shahepu and Lingyansi) (Fig. 1). To ensure the comprehensiveness and accuracy of the data, the data of the official real-time daily air quality reports and Ground-based measurements of air pollutants in this megacities were derived from different third-party sources [87, 88] . For example, the main pollutants come from National Urban Air Quality Real-time Publishing Platform, China air quality monitoring platform. The Environmental Quality Report released by Sichuan Environmental Monitoring Center and Chengdu Environmental Protection Busreau also were reviewed. The parallel time series of daily meteorological data were achieved from the China Meteorological Data Service Center (CMDC), Chengdu Meteorological Monitoring Database. Besides, the basic geographic information data and spatial administrative boundaries of the study area were obtained from the Geospatial Data Cloud and National Basic Geographic Information Center [89, 90]. Additionally, socioeconomic data were obtained from the National Bureau of Statistical of China (NBSC, 2021), Sichuan and Chengdu Statistical Yearbook. Missing data is completed by referring to the corresponding regional data.

2.3. Analysis Methods

In this study, the main reference for air quality evaluation is the Chinese standard (HJ 633-2012) (Table S3). Statistics on the exceeding standard of pollutants were based on the CAAQS and the latest international standards (Table S4). The validity of pollutant concentration was tested based on relevant Chinese national standards like GB3095-2012, HJ 663-2013, GB/T8170-2008 and HJ 194-2017 [23, 91]. Besides, missing data were processed with monitoring data from neighboring stations through a Kriging interpolation method. Finally, the data were checked by comparison with historical data. Excel, Origin, SPSS, and ArcGIS were used for spatial analysis of population, GDP, and topography, and to test correlations between PM and gaseous pollutants. The main pollutants were discussed by linear regression analysis.

3. Results and Discussions

3.1. Variation Characteristics of Pollutants

3.1.1. Annual variation

Tan et al. [60] analyzed the multi-temporal and spatial distribution characteristics of air quality in the Chengdu-Chongqing region from 2015 to 2021 and their results showed that the air pollutant concentration and AQI showed a downward trend year by year [60]. From 2014 to 2020, the air quality compliance rate in Chengdu also showed an upward trend. Compared with 2014 (61.1%), 2021 (81.96%) has increased by 34.14% (Fig. 2), indicating that the air quality in Chengdu has improved significantly. In addition, PM concentrations ($PM_{2.5}$: 77 μg/m^3, PM_{10}: 123 μg/m^3) in 2014 decreased by 76.48% and 50% respectively compared with 2021. This is consistent with the research results of Li et al. [92], whose research also pointed out that PM concentration has shown a downward trend in recent years in Chengdu [92]. SO_2 and CO also showed a sharp downward trend, down 68.4% and 50% respectively compared to 2014. The sharp reduction of SO_2 and CO shows that Chengdu has implemented a tough battle for clean energy alternatives, and has achieved remarkable results in strengthening

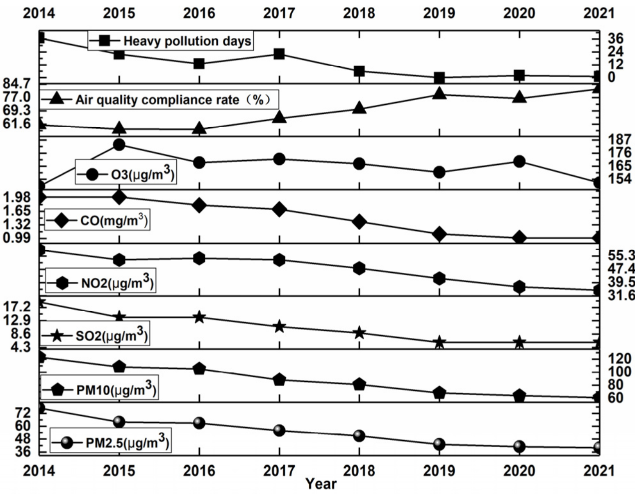


Fig. 2. Chengdu's air quality from 2014-2020.

industrial and coal-fired pollution control [93-95]. Contrary to other pollutants, O_3 showed a fluctuating growth, with an increase of 0.02% in 2021 (151μg/m^3) compared with 2014 (148μg/m^3). The research results of Wang et al. [96] also pointed out that O_3 in Chengdu showed a trend of fluctuating growth. During the "13th Five-Year Plan period" (2016-2020), the concentration of pollutants decreased significantly, and the annual average concentrations of SO_2, NO_2, PM_{10}, and $PM_{2.5}$ droped by 57.0%, 35.19%, 41.9% and 34.9%, respectively [96]. This showed that the air quality had improved from 2016 to 2020, which is consistent with the findings of Tan et al. [60]. This is mainly contributed to the implementation of measures such as the "650" project for air pollution control, the summer ozone control action plan, the implementation plan for Chengdu to win the battle to defend the blue sky, and the Chengdu air quality compliance plan (2018-2027) (Table S2) [52-60]. In 2020, the "636" project of low-carbon city construction was implemented in depth [97, 98]. The proportion of days with air quality up to standard in each district (city) county ranges from 72.7% to 91.8%. During the study period, there were certain differences in the concentration of each pollutant at the 8 monitoring sites (Fig. S7). Kuang et al. [99] analyzed the pollutant concentration data from 23 air quality monitoring stations in Chengdu, and the results also found that pollutant concentrations differed at each sites. $PM_{2.5}$ and PM_{10} maximums were mainly found at Junpingjie and Jinquanlianghe sites [99]. In addition, the value of O_3 at Lingyansi station in 2014-2020 is about 1.5-1.7 times that of WHO, 2021AQGs and CAAQS Grade-I (100μg/m^3). The maxima of SO_2 and NO_2 appeared at Lingyansi and Sanwayao sites, respectively. The difference in the value of pollutants at each site further indicates that there are differences in local pollution emissions in each region. When formulating pollution reduction measures, targeted local reductions should be carried out according to the pollution characteristics of each station.

3.1.2. Seasonal variation

During the study period, $PM_{2.5}$ and PM_{10} showed a "U-shaped" seasonal variation, with the lowest values in summer and the peak in winter (Fig. 3). In addition, $PM_{2.5}$ and PM_{10} showed a downward trend from 2014 to 2020, which shows that the "Sichuan Province Blue Sky Defense Action Plan (2017-2020)" and "Joint Prevention and Control Work System for Air Pollution Prevention and Control in the Chengdu Plain Area" have achieved remarkable results [64, 100]. However, the $PM_{2.5}$ concentration is still far higher than the WHO, 2021AQGs limits, especially in winter. For example, the $PM_{2.5}$ value in winter of 2017 was about 7 times that of CAAQS Grade I standard (15μg/m^3) and 20 times that of WHO, 2021AQGs (5μg/m^3). Relevant studies have shown that unlike cities in northern China, mobile sources (such as vehicles) and stationary sources (e.g., industries) are the main contributors to the higher $PM_{2.5}$ concentrations in this region in winter [99, 101-102]. Furthermore, O_3 showed an inverted "U-shaped" variation, with the peak value in summer and the lowest value in winter. This is consistent with the findings of Tan et al. [60], and they also observed that O_3 showed an inverted "U-shaped" seasonal change during their study [60]. In summer, strong solar radiation and high temperature can promote the tropospheric photochemical reaction rate, thereby increasing the conversion rate between O_3 and precursors and producing abundant O_3 [103]. On the contrary, gaseous pollutants NO_2, CO, and SO_2 showed a "U-shaped" seasonal variation. The concentration of NO_2 is still large, with the annual value being about 3-5 times that of WHO, 2021AQGs (10μg/m^3), indicating that NO_2 is still the main pollutant that needs to be reduced in the future. Emissions are an internal factor affecting the seasonal variation of pollutants, while external factors such as unfavorable meteorological conditions are also major contributors [38, 104-106]. Due to its closed terrain, Chengdu is prone to static and stable weather in winter and pollutants are easy to accumulate. The seasonal difference in the pollutants also indicates that different measures should be taken in different seasons. For example, during the summer when ozone pollution is severe, the focus is on strengthening the control of VOCs emitting companies and implementing peak-shifting production [107]. In addition, classified management is implemented for key industries such as petrochemical, chemical, and automotive painting. In winter, when $PM_{2.5}$ pollution is serious, pollution prevention and control in cement, brick and tile enterprises will be strengthened.

3.1.3. Monthly variation

By analyzing the fluctuation characteristics of pollutants between adjacent stations, it can provide a basis for tracing their source direction. The steeper the peak shape, which indicates shorter durations, the greater the difference between adjacent sites, and the more likely a local emission effect. From 2014 to 2020, the change trend of min AQI, max AQI and average AQI in Chengdu is the same. Overall, the Max AQI ranged from 50 to 150, and the air class was moderate for most months during the study period (Fig. 4). The average AQI is in the range of 50-100 except for individual months with values higher than 100, indicating that the overall air quality during the study period was moderate. Wang et al. [96] analyzed the spatial and temporal trajectory evolution and causes of air pollution in Chengdu and also found that the air quality in Chengdu was moderate during the study period (2015-2018) [96]. There were obvious monthly fluctuations in $PM_{2.5}$ and PM_{10} at each site during the research period, indicating that PM is affected by local emissions (Fig. S8). The findings of the

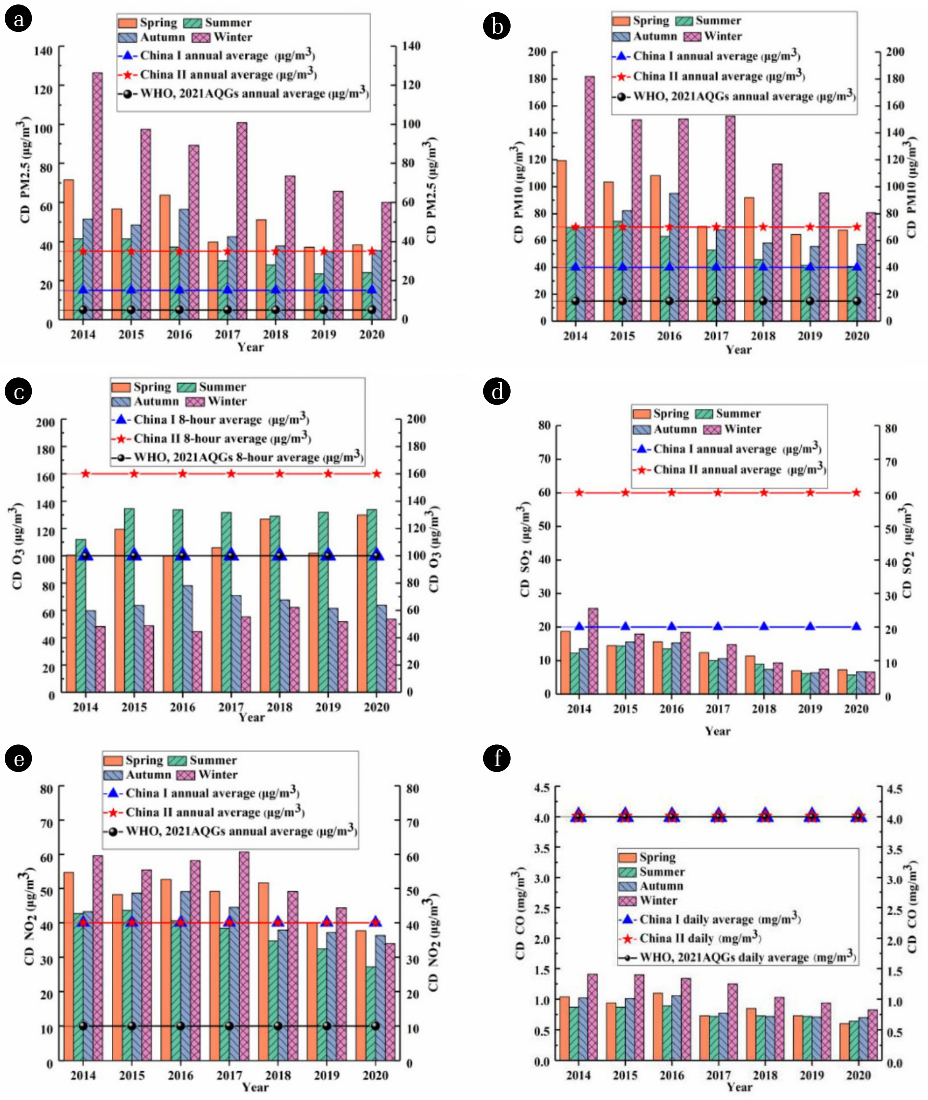


Fig. 3. Seasonal changes of major pollutants from 2014 to 2020: (a) $PM_{2.5}$, (b) PM_{10}, (c) O_3, (d) SO_2, (e) NO_2, (f) CO

research by Kong et al. [108] also pointed out that air pollutants in the Chengdu showed significant monthly (seasonal) differences [108]. In 2020, the main sources of PM were industrial emissions (61.63%), followed by domestic emissions (38.33%) [109]. The monthly variation trend of O_3 is also obvious, which is consistent with the findings of other scholars, both showing that it is higher from January to February and lower from June to August [96, 110]. In addition, the O_3 concentration value is lower in the month with high PM value, indicating that the reduction of PM concentration can promote the generation of O_3 to a certain extent. Therefore, emission reduction measures need to consider the synergistic effect of $PM_{2.5}$ and O_3. Tan et al. [60] also found that O_3 values were higher from June to August and lower from January to March and October to December [60]. In addition, the monthly changes in CO and SO_2 remained stable, indicating that the differences between sites were not significant. The monthly variation of NO_2 at each site is more obvious (Fig. S9), and its main sources are industrial sources (61.63%) and domestic sources (like transportation sources, etc.) [109].

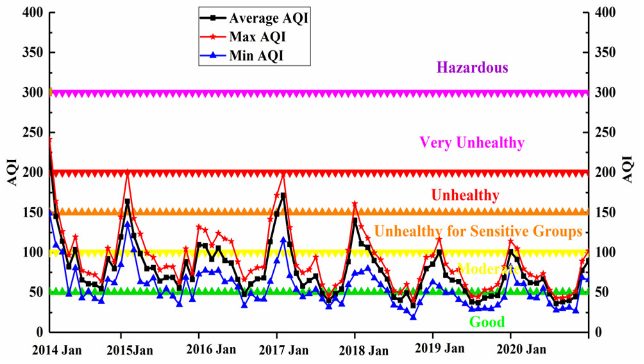


Fig. 4. Monthly trend of AQI between 2014-2020.

3.2. Attainment of Air Quality Standards

During the study period, the air quality level improved significantly, and the AOI value in 2020 (60) decreased by 42.31% compared with 2014 (104). In 2020, the proportion of days with good air quality was 76.5%, an increase of 9.6% over 2015 [109]. This may be due to the lower contribution of traffic and industrial pollution during the COVID-19 lockdown period [111, 112]. Xia et al. [113] also observed that the lockdown measures during COVID-19 help improve the air quality in Chengdu [113]. However, the PM concentration value is still far greater than the CAAQS standard, and is far from the WHO, 2021AQGs standard. The annual average concentrations of PM at each station have exceeded the CAAQS standard ($PM_{2.5}$: 15 μg/m^3, PM_{10}: 40 μg/m^3) for 7 consecutive years, which are higher than the WHO, 2021AQGs standard. The concentration of $PM_{2.5}$ (81μg/m^3) in 2014 was 16 times that of the WHO, 2021 AQGs standard (5μg/m^3), and 5-6 times that of the CAAQS Grade I standard (15μg/m^3). For PM_{10}, the concentration value in 2020 was significantly reduced by 45.5% compared with that in 2014 (Fig. 5). Particulate matter is still an important target for future air pollutant control in Chengdu, especially $PM_{2.5}$.

Compared with 2014 (17.6μg/m^3), the SO_2 concentration value in 2020 (6.6μg/m^3) has dropped by about 62.5%, which is mainly due to the mitigation of the use of non-fossil fuels in Chengdu and the vigorous development of clean energy [96]. The annual average concentration of NO_2 from 2014 to 2018 was higher than the CAAQS standard (40μg/m^3), and still had a large gap from the WHO, 2021AQGs standard (10μg/m^3). The NO_2 emission in Chengdu is mainly from mobile sources, and the number of civilian vehicles in 2020 has increased by 77.85% compared with 2014. The study by Zhou et al. [90] also revealed that traffic sources are the main contributor to NO_2 in Chengdu [90]. By detrended cross-correlation analysis (DCCA), Shi et al. [114] found that NO_2 concentration fluctuations are positively correlated with urban traffic congestion in the form of a power function in Chengdu [114]. In addition, the O_3 problem has gradually become prominent. The research of Tan et al. [115] also pointed out that reducing VOCs is the most effective way to alleviate ozone pollution in Chengdu [115]. On the one hand, NO_x and VOCs emitted by mobile sources are the main precursors of O_3 and PM, and on the other hand, the emission reduction of PM promotes the generation of photochemical O_3 [116-120]. Through the discussion of VOCs and NOx reduction schemes, Chen et al. [70] suggested that Chengdu was typical in the VOC-limited regime, and reducing VOCs emissions is the key to the prevention and control of O_3 [70]. Therefore, NO_x emission control pattern should be carefully mapped to assess changes in O_3. In 2020, the coal-fired boilers in Chengdu have been cleared, and the proportion of clean energy rise from 56.5% in 2015 to 62.6% in 2020 [121]. O_3 is the third largest greenhouse gas after CO_2 and CH_4 [122, 123]. In the future pollution control work, it is particularly important to strengthen the research on O_3 pollution and its emission reduction.

3.3. Correlations Between Air Pollutants

3.3.1. $PM_{2.5}/PM_{10}$ ratio

Exploring the change in the proportional relationship of different pollutant concentrations can determine the source or type of pollutants, which helps to improve the accuracy of pollution control. $PM_{2.5}$ and PM_{10} have different physical and chemical properties, and previous studies have revealed that the ratio of $PM_{2.5}/PM_{10}$ can indirectly provide some indicators, such as pollutant composition, source contribution, and impact on health [124-128]. Overall, a lower $PM_{2.5}/PM_{10}$ ratio demonstrate a predominance of PM_{10} (mainly from natural sources), and a higher ratio indicates more air pollution from anthropogenic sources [124-126]. Compared with other cities in China, the PM_{10} and $PM_{2.5}$ levels in Chengdu were bigger than those in Beijing and Guangzhou [67, 129].

Combined with previous studies, the year-on-year declines in $PM_{2.5}$ and PM_{10} observed in this study are mainly due to strict air quality control policies and other measures [82, 108, 130]. However, the mean value of the $PM_{2.5}/PM_{10}$ ratio in Chengdu ranged from 0.6 to 0.65 (Fig. S10), indicating that PM pollution is still serious [80, 131]. Qi et al. [121] pointed out that the greater the ratio, the higher the risk to public health [121]. PM mainly comes from the construction, cement industry and automobile exhaust. Therefore, precursors should be reduced, and the use of high-emission vehicles should be banned. The monthly $PM_{2.5}/PM_{10}$ ratios are not constant, high in winter and low in summer (Fig. S11). However, the ratio in different seasons did not exceed the range (0.5-0.8) of urban areas in developed countries. The change of $PM_{2.5}/PM_{10}$ ratio was due to the different dominant sources and their contribution rates in different seasons [121, 129]. Furthermore, this ratio fluctuates relatively steadily from March to September every year, and it is about 0.58 between 2014 and 2020 (Fig. S12). Obviously, the ratio of $PM_{2.5}/PM_{10}$ is constantly fluctuating, indicating that urban air pollution is not a single source of pollution. Therefore, the effective control and management of PM is particularly critical.

3.3.2. NO_2/SO_2 ratio

NO_2 mainly comes from stationary (burning) and mobile sources (vehicle exhaust)[132-134]. While, SO_2 is a traditional industrial pollutant mainly derived from stationary sources such as coal combustion, power generation and industrial production [135-139]. The energy structure characteristics of a region can be reflected by the mass concentration ratio of NO_2 and SO_2 [140, 141]. At the same time, the ratio also can reflect the regional coal suppression and gas desulfurization effects, as well as the relative changes in the characteristic pollution of automobile exhaust. By analyzing the change of NO_2/SO_2 ratio, it is helpful to identify the main sources of regional pollutants. A higher value of this ratio indicates that the pollutants are from mobile sources, and conversely, it indicates that the stationary sources are higher.

From 2014 to 2020, the effect of SO_2 emission reduction was obvious, while NO_2 decreased with fluctuations. Overall, the NO_2/SO_2 ratio showed a clear upward trend (Fig. S13). This is consistent with the observes of Zhao et al. [142], who found that the NO_2/SO_2 ratio in Chengdu in 2018 (5.12) showed a significant growth trend compared to 2008 (1.06) [142]. In addition, compared to 2014 (2.85), the ratio surged up to 5.15 in 2020, an increase of 80.7%. A large number of studies have shown that vehicles are the main source of NO_x in megacities [143-147]. In 2016, NO_x emissions from motor vehicles accounted for 50% of Chengdu's

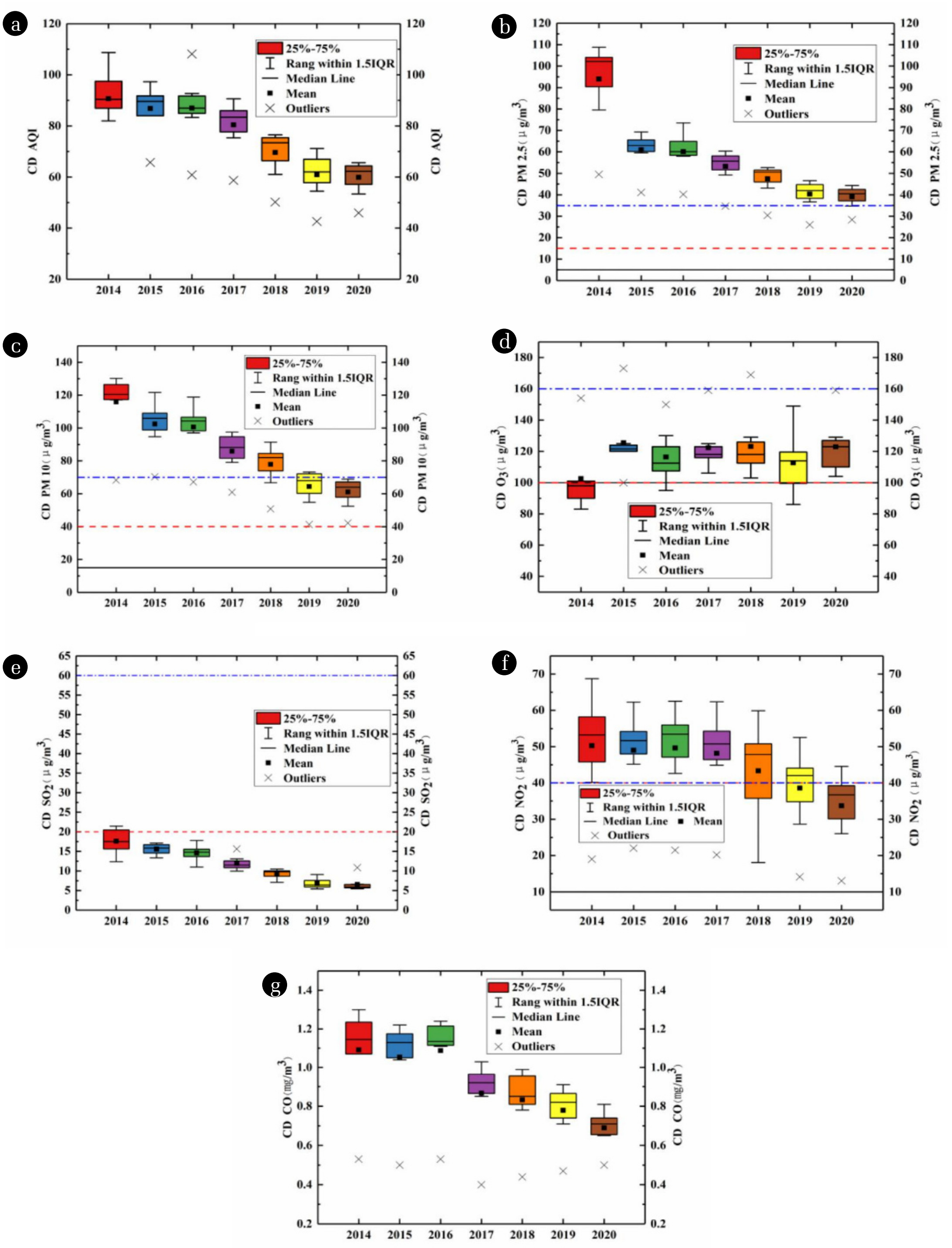


Fig. 5. The average annual of AQI and six criteria air pollutants in Chengdu (The red, blue and black dotted lines show the daily and annual limits specified by CAAQS Grade I, CAAQS Grade II standard and WHO, 2021AQGs, respectively): (a) AQI, (b) $PM_{2.5}$, (c) PM_{10}, (d) O_3, (e) SO_2, (f) NO_2, (g) CO

NO_x emissions. Compared with 2014, Chengdu's population and car ownership increased by 29.32% and 77.85% respectively in 2020. The sharp increase in the number of vehicles and the increase in human activities are considered to be the main reasons for the increase in the NO_2/SO_2 ratio. However, compared with 2019 (5.69), this ratio showed a certain downward trend in 2020 (5.15). The main reason is that due to the impact of the COVID-19 epidemic in 2020, most cities in China adopted lockdown measures, resulting in a reduction in NO_x emissions from industry and transportation [141, 148-150]. Zhang et al. [82] found that NO_2 concentrations decreased 47.0% during the COVID-19 lockdown in Chengdu [82]. Overall, the gradual increase in the NO_2/SO_2 ratio also indicates that mobile sources of pollution such as vehicle exhaust emissions have a higher contribution than stationary sources such as coal combustion, power generation, and industrial production [18]. In addition, the NO_2/SO_2 ratio showed an increasing trend in each season (Fig. S14). Although desulfurization equipment is widely used in industry and power plants, the emission reduction of SO_2 and NO_x is large, but the dramatic rise in the number of vehicles and total electricity consumption greatly offsets the reduction effect of NO_x. The NO_2/SO_2 ratio varies greatly from month to month. Zhao et al. [142] found that the monthly average NO_2/SO_2 concentration in the commercial and industrial areas was relatively large, indicating that factors such as industrial structure, traffic, and population had a greater impact on NO_2 and SO_2 pollution emissions [142]. Some study also showed a significant positive correlation between NO_2/SO_2 ratio and mortality [18, 151]. In addition, high concentrations of SO_2 and NO_2 will accelerate $PM_{2.5}$ pollution [18, 151]. Therefore, reducing NO_2 emissions is particularly necessary for pollution prevention and health.

3.3.3. Correlations between air pollutants

There are complex connections between various types of air pollutants, and exploring the correlation between air pollutants is critical to further improve the accuracy of pollution prevention and carry out air pollution control. The mass concentration of PM_{10} and $PM_{2.5}$ showed an obvious positive correlation, and the correlation coefficient reached 0.978 (Table 1). This is consistent with the research results of some scholars [60, 152]. In addition, the seasonal correlation coefficients were all greater than 0.9, indicating that the seasonal correlation between $PM_{2.5}$ and PM_{10} was extremely strong. The possible reason is that $PM_{2.5}$ is a component of PM_{10}, and they have similar sources and pollution transformation laws [18, 153]. There is an obvious linear correlation between $PM_{2.5}$ and PM_{10}, the correlation coefficient R^2=0.930, the regression equation is: $PM_{2.5}$=-3.34+0.66PM_{10}, R=0.964 (Fig. S15). It further shows that the linear correlation between them is extremely strong, and the regression effect is better. It also can be speculated that there is a more complex relationship between $PM_{2.5}$ and PM_{10}.

There was a significant inverse correlation between PM and O_3 ($PM_{2.5}$ and O_3: R = -0.552; PM_{10} and O_3: R = -0.509), and the correlation was moderate. This is similar to the findings of the study by Kuang et al. [99], Which also pointed out that the increase of PM concentration can slow down and inhibit the generation of O_3 to a certain extent [99]. In addition, their study also found that O_3 pollution in central Chengdu is mainly caused by vehicle emissions, and that industries in the northern region have a more important impact on O_3. PM was positively correlated with SO_2, NO_2 and CO, with correlation coefficients ranging from 0.73 to 0.89, indicating that they are highly homologous (e.g., fossil fuel combustion). Studies have shown that reducing emissions of gaseous pollutants such as SO_2, NO_x and NH_3 leads to lower concentrations of particulate pollutants [2]. In addition, O_3 was negatively correlated with SO_2 and CO, but positively correlated with NO_2 (R=0.428). Primary pollutants such as NO_2 emitted by human activities can effectively accelerate the formation of O_3 through photochemical reactions. A large number of studies have shown that controlling NO_x and VOCs is an effective way to reduce O_3 [115, 154-157]. For example, Tan et al. [115] analyzed the sensitivity of O_3 to its precursors through an observation-based box model and found that the relative incremental reactivity of OVOCs was greater than that of other precursors, indicating that OVOCs also played a dominant role in O_3 formation [115]. There was a significant positive correlation between SO_2, NO_2, and CO, indicating that the three pollutants were homologous, mainly from fossil fuel combustion and vehicle exhaust emissions [158]. There are also different correlations between pollutant concentrations in different seasons. Except for O_3, each pollutant showed a strong positive correlation (R>0.8) in the four seasons. The correlation between NO_2 and PM is larger in summer, indicating that atmospheric particulate matter is more sensitive to NO_2 changes in summer [18]. In the future emission reduction, Chengdu should focus on industrial sources, mobile sources, dust sources, etc., to advance the coordinated emission reduction of multi-pollutants, and further reduce the concentrations of $PM_{2.5}$ and O_3. For example, strengthen the governance of key areas, key periods, and key industries, and implement synergistic emission reductions of NO_x and VOCs.

Table 1. Correlations of pollutants in Chengdu based on monthly data in 2014-2020

Pollutants	PM_{10}	SO_2	NO_2	CO	O_3
Yearly					
$PM_{2.5}$	0.978	0.732	0.810	0.877	-0.552*
PM_{10}		0.778	0.864	0.890	-0.509
SO_2			0.778	0.829	-0.252
NO_2				0.791-	0.428
CO					-0.536
Spring					
$PM_{2.5}$	0.991	0.925	0.821	0.936	-0.349
PM_{10}		0.920	0.824	0.933	-0.267
SO_2			0.901	0.879	-0.442
NO_2				0.835	-0.373
CO					-0.502
Summer					
$PM_{2.5}$	0.990	0.949	0.931	0.925	-0.415
PM_{10}		0.949	0.938	0.911	-0.337
SO_2			0.948	0.926	-0.181
NO_2				0.908	-0.352
CO					-0.325
Autumn					
$PM_{2.5}$	0.996	0.948	0.887	0.967	0.426
PM_{10}		0.955	0.879	0.981	0.361
SO_2			0.957	0.963	0.315
NO_2				0.863	0.493
CO					0.228
Winter					
$PM_{2.5}$	0.969	0.954	0.854	0.894	-0.414
PM_{10}		0.958	0.939	0.959	-0.439
SO_2			0.820	0.934	-0.606
NO_2				0.915	-0.339
CO					-0.561

3.4. Coordinated Control of Greenhouse Gases and Pollutants

3.4.1. CO_2 emissions

At present, the coordinated management of greenhouse gases and air pollutants has become an urgent need for urban environmental management [159]. China faces the dual challenges of conventional air pollution control and greenhouse gas emission reduction. In 2020, China's total energy consumption reached 498,000 (10,000 tce), an increase of about 2.39 times compared to 2000 (Fig. S16). Miao et al. [160] also calculated that China's total CO_2 emissions increased from 9.122 billion tons in 2011 to 9.912 billion tons in 2020 [160]. Although the use of coal has decreased and the development and utilization of clean energy has increased, the overall structure of China's use of coal as the main energy source has not changed [161]. Coal accounts for 67% of total energy consumption annually. This has resulted in 90%, 67%, 70% and 70% of China's SO_2, NO_x, PM, and CO_2 emissions from burning coal, respectively. From 2002 to 2013, CO_2 emissions were in a rapid growth stage, mainly due to the rapid development of domestic steel, cement, and other heavy industries. The emissions rose at an average annual rate of 9.6%, making it the world's largest emitter. From 2014 to 2020, the growth rate slowed down, and the emission decreased slightly, which may be due to the contribution of energy-saving renovation in the industrial industry, but the growth trend resumed in 2017 (Fig. 6). The total CO_2 emissions in 2020 were 10,591.37 million tones (Mts), accounting for 27% of the global total, an increase of about 2.30 times compared to 2000 (3,214.07 Mts). Coal is the main source of CO_2 emissions, occupied more than 75% of the China's total emissions. Zhang et al. [162] also pointed out that reducing the use of coal is the key path to achieve carbon neutrality [162]. It notes that the natural gas total emission shows a dramatic jump, and the emission in 2020 (415 Mts) is 10.86 times that in 2000 (38.20 Mts).

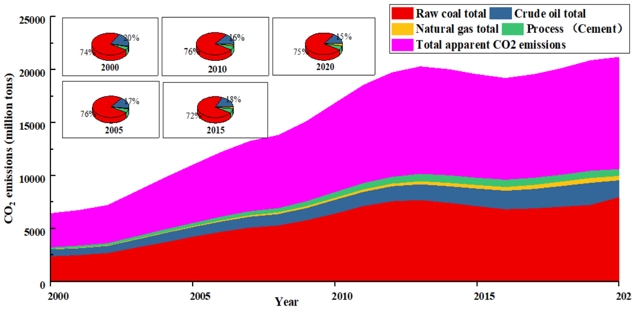


Fig. 6. China's CO_2 emissions (2000-2020)

There are regional differences in CO_2 emissions in mainland China [160]. In 2019, the 30 provinces in China with the largest carbon emissions were Shandong (745.57 Mts), followed by Jiangsu (419.5 Mts) and Hebei (409.11 Mts) [25]. Emissions from Sichuan province reached 167.75 Mts. All provinces and cities accounted for the largest proportion of raw coal emissions, and Sichuan Province accounted for 80.78%. Yang et al. [97] found that the increase in energy consumption is the main reason for the growth of CO_2 emissions in Chengdu [97]. In general, the areas with the largest carbon emissions are concentrated in the central and eastern parts of China with developed industries and agglomerated populations. This is consistent with the research results of Qian et al. [163], whose research also shows that the growth of China's CO_2 emissions is mainly concentrated in urban agglomerations [163]. The trend of carbon emissions in Sichuan Province is remained stable (90-113 Mst), with an average annual emission of about 262 Mts. The research by Cui et al. [164] revealed that the total CO_2 emissions in cities such as Chengdu and Chongqing present a high-low clustering pattern [164]. From 2007 to 2012, Chengdu's carbon emissions display an increasing trend, which may be related to the rapid development of industries. There was a certain downward trend in 2012-2015, but it rose sharply in 2015-2019 and climbed to a high point (59.7Mts) in 2019 (Fig. S17). Li et al. [165] also pointed out that Shenzhen, Chengdu, and Guangzhou have higher CO_2 emission levels [165]. Yang et al. [97] found that the increase in energy consumption is the main reason for the growth of CO_2 emissions in Chengdu [97]. Their research also pointed out that striving to stimulate more low-carbon potential and momentum and strengthen green transformation in all aspects of production and life are the key to helping Chengdu achieve carbon peak. Chengdu's carbon emissions take up 12.5%-21.6% of the total emissions in Sichuan Province, which indicates that this megacity's carbon emission reduction plays a key role in promoting the realization of the carbon peak in Sichuan Province, and at the same time promotes the construction of the Cheng-Yu economic circle.

3.4.2. "Pollution and carbon reduction" synergistic effect

Greenhouse gases and atmospheric pollutants have the same origin. Air pollution and climate change affect each other, air pollution can cause climate effects, and climate change affects the diffusion and transformation of air pollution to a certain extent [166, 167]. In general, climate change and air pollution control have a high degree of synergy in terms of scientific mechanisms, target indicators, countermeasures, comprehensive benefits and governance systems [168-170].

Relevant studies show that from 2015 to 2020, the industrial sector has achieved a 6% reduction in CO_2 emissions, and the major pollutants SO_2, NO_x and $PM_{2.5}$ have been reduced by 56%, 19%, and 37%, respectively [25]. But the magnitude of greenhouse gas emissions is much higher than that of atmospheric pollutants. In addition to the industrial sector, the power, heating, civil and transportation sectors continued to increase their CO_2 emissions while the emissions of major air pollutants declined. Among them, the emission reduction of pollutants in the power and heating sectors is mainly based on terminal control measures, and it is impossible to achieve coordinated emission reduction of CO_2. During the China Blue Sky Defense War (2018-2020), a large number of measures were taken, and the synergistic effect of $PM_{2.5}$ and CO_2 emission reduction was obvious (Fig. 7) [171]. It shows that a series of major measures in China's industrial restructuring in recent years (elimination of outdated production capacity, renovation of industrial boilers, comprehensive renovation of scattered and polluting enterprises, etc.) have achieved good results. The clean substitution of bulk coal has achieved initial results in the coordinated reduction of CO_2 emissions. Overall, CO_2 emissions have the same source as SO_2, NO_x and $PM_{2.5}$ and other air pollutants

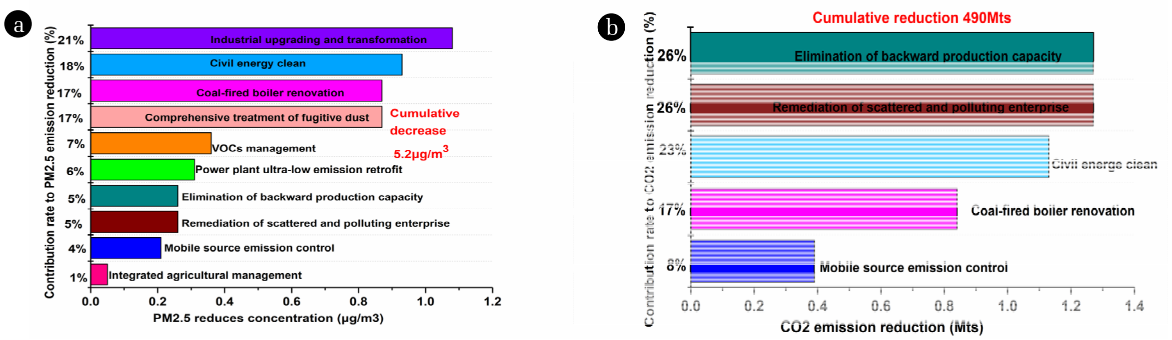


Fig. 7. Coordinated emission reduction of main measures in the Blue Sky Defense War: (a) $PM_{2.5}$ reduces concentration, (b) CO_2 emission reduction

[166, 167]. Implementing CO_2 emission reduction measures in industry can significantly reduce pollutant emissions. Continuous improvement of ambient air quality and dual-carbon governance actions are important means for synergistic effect of pollution control and carbon reduction. The reduction potential of air pollutants in China's energy, industrial and transportation structure adjustment needs to be further released, and the next step should be to actively promote the source emission reduction measures to achieve the synergistic effect of pollution and carbon emigration [168, 172]. From 2014 to 2020, Chengdu's CO_2 emissions showed a certain growth trend, and reached the maximum (60Mts) in 2019 (Fig. S17). Except for O_3, which showed an increasing trend, the rest of the pollutants showed a decreasing trend. It shows that the reduction potential of air pollutants still needs to be improved, and it should be promoted synergic reduction of pollution and carbon.

3.4.3. Coordinated control of CO_2 and pollutants

Climate change and air pollution are highly homologous and have complex correlations [26, 40, 173-177]. The "dual carbon" goal has become an important driver for coordinating high-quality economic and social development and high-level protection of the ecological environment. Building materials, steel, electricity and motor vehicles are the main contributors to greenhouse gases and $PM_{2.5}$-related pollutants in Chengdu. Chengdu should focus on the "dual carbon" goal, focusing on energy, industry, construction, transportation and other fields, and propose a roadmap for collaborative control technology. At the same time, the heterogeneity of pollutants and greenhouse gases should be fully identified, so as to deepen the coordinated reduce of greenhouse gases and atmospheric pollutants.

Changing the energy structure dominated by fossil energy is the core way to achieve carbon neutrality and also the key to reducing pollutant emissions [97, 178]. After 2030, as the emission reduction potential of end-of-pipe governance measures decreases, the deep low-carbon energy transformation measures under the carbon neutrality goal will become the driving force for continuous and deep improvement of air quality. Therefore, giving priority to coordinated emission reduction measures to reduce the use of fossil fuels in the process of policy formulation is the main way for Chengdu to achieve synergies in pollution reduction and carbon reduction [97]. More importantly, the energy transition under the "dual carbon" goal will also bring huge health benefits. Relevant studies show that 1,120 $PM_{2.5}$-related premature deaths can be avoided when China's power sector decarbonizes and reduces emissions by 195 million tons of CO_2 [25]. Chengdu needs to greatly increase the scale of wind power and photovoltaic power generation and promote the expansion of the input channels of hydropower in western Sichuan and photovoltaic power in northwestern Sichuan. In addition, the emission of industrial process pollutants such as iron and steel, cement, metallurgy, building materials and petrochemicals is a critical area of air pollution control in Chengdu at present, and its process carbon emission is also a difficulty in future emission reduction. In the short term, the greening level of the industry should be improved by means of energy efficiency improvement and fuel structure optimization, and the emission reduction potential should be continuously released. Different studies have also pointed out that traffic pollution in Chengdu is still relatively serious [90, 179-182]. The transformation of the traffic structure will further reduce CO_2 emissions and improve air quality, while bringing huge health and economic benefits [167, 183]. Overall, the deep emission reduction of CO_2 cannot only rely on the adjustment of energy, transportation and industrial structure, but also should pay attention to the research and development of emerging low-carbon, zero-carbon or negative-carbon technologies.

Conclusions

Scientific emission reduction in complex terrain areas under the goal of coordinated environmental and climate governance is challenging. Carbon neutrality goal can speed up O_3 and other pollution control while promoting carbon emission reduction. The main research conclusions and suggestions of this study are as follows:

(1) From 2014 to 2020, the concentrations of $PM_{2.5}$, PM_{10}, SO_2 and CO have dropped significantly, but $PM_{2.5}$ and PM_{10} still far exceed the limits of CAAQS and the latest WHO, 2021AQGs standards.

(2) The peaks of $PM_{2.5}$ and O_3 have been stagnant in the past few years, becoming important pollutants for future air pollution control in Chengdu, and refined mitigation strategies should be sought.

(3) The O_3 concentration at most stations exceeded the standard

and showed a fluctuating upward trend. In addition, O_3 showed an inverted "U-shaped" seasonal variation and there is a "seesaw" phenomenon between O_3 and PM. This indicates that priority control sources should change accordingly based on the season.

(4) There are complex correlations between pollutants, and at the same time, they are homologous to greenhouse gases, and emission reduction measures should consider multi-pollutant synergy.

Continuous improvement of ambient air quality and dual-carbon governance actions are important means for synergistic effect of pollution and carbon reduction. Chengdu should focus on the "dual carbon" goal and deepen the coordinated control of greenhouse gases and air pollutants from the aspects of energy, industry, transportation structure and zero-carbon negative carbon technology. At the same time, the coordinated control of $PM_{2.5}$ and O_3 is the main line, and the coordinated emission reduction of multi-pollutants is advanced. Emission reduction strategies need to comprehensively consider their air quality health benefits and climate effects, strengthen differentiated collaborative management, and control, and implement dynamic management. In addition, the relationship between regional air quality, land use, industrial structure, pollution sources, meteorological elements and the transport and transformation of pollutants needs further investigation. Research should also be conducted at finer spatial and temporal scales, incorporating more novel approaches to compare and validate multiple simulation methods. Finally, the multi-goals of "energy-environment-health-climate" should be comprehensively considered, focusing on the coordinated control of $PM_{2.5}$ and O_3, the joint emission reduction of pollutants and greenhouse gases, and the risk management and control of harmful pollutants.

Conflict-of-Interest Statement

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Author Contributions

X.J.L. (PhD student) performed all data simulations and analyses, and wrote the manuscript. L.C.A. (Professor) supervised and revised the manuscript. S.S. (Doctor) revised the manuscript. M.S.M.S. (Doctor) revised the manuscript. S.A.H. (Associate Professor) revised the manuscript. T.P.A. (Doctor) revised the manuscript. J.Z.H. (Professor) revised the manuscript.

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