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Comparative Analysis of Long-Term Variation Characteristics of SO₂, NO₂, and O₃ in the Ecological and Economic Zones of the Western Sichuan Plateau, Southwest China

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Abstract: Sulfur dioxide (SO₂), nitrogen dioxide (NO₂), and ozone (O₃) are important atmospheric pollutants that affect air quality. The long-term variations of SO₂ and NO₂ in 2008–2018 and O₃ in 2015–2018 in the relatively less populated ecological and economic zones of Western Sichuan Plateau, Southwest China were analyzed. In 2008–2018, the variations in SO₂ and NO₂ in the ecological zone were not significant, but Ganzi showed a slight upward trend. SO₂ decreased significantly in the economic zone, especially in Panzhihua, where NO₂ changes were not obvious. From 2015 to 2018, the concentration of O₃ in the ecological zone increased significantly, while the economic zone showed a downward trend. The rising trend of the concentration ratio of SO₂ to NO₂ in the ecological zone and the declining trend in the economic zone indicate that the energy consumption structure of these two zones is quite different. The lower correlation coefficients between NO₂ and O₃ in the Western Sichuan Plateau imply that the variations of O₃ are mainly affected by the regional background. The effects of meteorological factors on SO₂, NO₂, and O₃ were more obvious in the economic zone where there are high anthropometric emissions.

Keywords: sulfur dioxide; nitrogen dioxide; ozone; Western Sichuan Plateau; long-term variation

1. Introduction

In recent decades, China's economy has developed rapidly. Especially in the 21st century, the emission of artificial air pollutants has increased significantly, and hazy weather is frequent; therefore, the air pollution problem has attracted people's attention. The Sichuan Basin is one of the areas in China suffering from serious air pollution due to its special weather and topography [1–4]. Sulfur dioxide (SO₂), nitrogen dioxide (NO₂), and ozone (O₃) are important pollutants that not only affect the generation of atmospheric fine particles and the photochemical and atmospheric acidification processes but also threaten human health [5–8]. Many previous studies [9–11] have carried out quantitative analysis of SO₂, NO₂, and O₃ pollutants, discussed the diurnal and seasonal variation characteristics, and pointed out that SO₂ and NO₂ have the lowest concentration in summer due to

the wet cleaning effect of precipitation, while O₃ has the highest concentration as a result of the most active photochemical reactions occurring in spring and summer, indicating that NO_x is an important precursor of O₃.

Several studies have focused on the variations of SO₂, NO₂, and O₃ in the most polluted areas in China, such as the Beijing-Tianjin-Hebei region, North China, and the Yangtze River and Pearl River Deltas. The results show that the concentration of SO₂ declined sharply in the past decade, especially after 2006, because of the SO₂ emission reduction requirement in China's Ninth Five-Year Plan (1996–2000). The concentrations of NO₂ in large and medium-sized cities of China showed a fluctuating upward trend due to the lack of effective denitration technology [12–17]. Due to the late observation time and complicated formation mechanism, few studies have been conducted on the long-term variation characteristics of O₃. In the past five years, serious O₃ pollution has appeared in the large cities of China, and O₃ has replaced PM_{2.5} as the primary pollutant in some cities [18–20]. Zhao et al. [21] analyzed the characteristics of six criteria air pollutants in the Sichuan Basin of Southwest China from 2015 to 2017 and pointed out that all air pollutants except O₃ showed U-shaped annual changes; motor vehicle emissions was the main contributor to O₃ in the basin, while industrial emission was the main contributor to O₃ in the Western Sichuan Plateau area. Air quality in many large and medium-sized cities has improved significantly as a result of various environmental policies implemented by the Chinese government [22,23]. The changes of air pollutants in the sparsely populated western plateau in the past decade remain in question.

The Sichuan Provincial Government [24] has designated Aba and Ganzi as ecological zones, and Liangshan and Panzhihua as economic zones based on the industrial structure and gross domestic product (GDP), respectively. They are located in a high-altitude area in the transition from the Sichuan Basin to the Tibet Plateau, called the Western Sichuan Plateau. Due to the sparsely populated land and low emissions of man-made pollutants, this is a clean air background region in southwest China and even the whole country. The study of the long-term variation characteristics of atmospheric pollutants in this region is of great scientific significance for a comparative analysis of pollutants in other regions of China. Based on the daily mass concentration of SO₂ and NO₂ from 2008 to 2018 and O₃ from 2015 to 2018, long-term variations and the basic characteristics of three gaseous air pollutants in the ecological and economic zones of Western Sichuan Plateau were compared and analyzed in this paper.

2. Materials and Methods

The ecological and economic zones of Western Sichuan Plateau are located east of the Tibetan Plateau. The ecological zone includes Aba and Ganzi, and the economic zone includes Liangshan and Panzhihua. The geographical locations of these zones are shown in Figure 1, the area outlined in blue is the ecological zone and the area outlined in red is the economic zone, and the locations of cities are marked by black dots. The average altitude of Aba, Ganzi, Liangshan, and Panzhihua is 3000 m, 4100 m, 1500 m, and 1500 m, respectively.

The air pollutants data used in this paper were collected from Sichuan Ecological Environment Monitoring Center (<http://www.scnewair.cn:6112/publish/index.html>) [25]. The citywide daily average concentrations of SO₂ and NO₂ from 1 January 2008 to 31 December 2018, and the citywide daily maximum eight-hour average concentration of O₃ from 1 January 2015 to 31 December 2018 were used to analyze the spatiotemporal distribution characteristics of three air pollutants in Western Sichuan Plateau. The mass concentrations of SO₂, NO₂, and O₃ were measured by the ultraviolet fluorescence method (TEI, Model 43i, Thermo Fisher Scientific Inc., Waltham, MA, USA), the chemiluminescence method (TEI, Model 42i, Thermo Fisher Scientific Inc., USA), and the UV spectrophotometry method (TEI, Model 49i, Thermo Fisher Scientific Inc., USA), respectively [21]. The measurements were conducted by multiple national air quality monitoring sites in cities [26]. The daily average concentrations of the three air pollutants released after quality assurance and control by the Sichuan Ecological Environment Monitoring Center based on Technical Guideline on Environmental Monitoring Quality Management HJ 630-2011 (<http://kjs.mep.gov.cn>) were the same as the daily air quality reports issued

by the government. In addition, at least 324 and 25 daily concentrations were required to calculate the average annual concentrations of SO₂, NO₂, and O₃.

The routine meteorological parameters were collected from the National Meteorological Information Center of China [27]. The surface stations are shown by black dots in Figure 1. The monthly mean meteorological parameters included mean monthly average temperature, maximum and minimum monthly average temperature, mean relative humidity, monthly total precipitation, monthly sunshine duration, mean wind speed, and maximum wind speed from January 2008 to December 2017. The data quality is controlled by the National Meteorological Information Center of China and the accuracy rate is close to 100%.

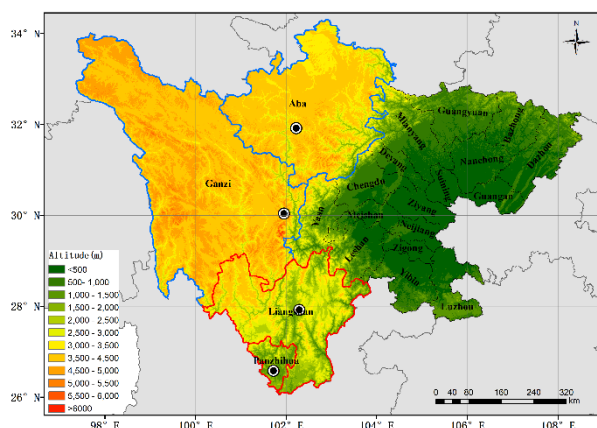


Figure 1. Geographical position of ecological zone (blue outline) and economic zone (red outline) of Western Sichuan Plateau, the locations of cities are marked by black dots.

The related statistical data of the basic situation of each city were collected from the statistical yearbook provided by the Sichuan provincial bureau (<http://tjj.sc.gov.cn/tjnj>). Table 1 shows the region category, area, resident population, urbanization rate, GDP, and possession of civil motor vehicles of the study area at the end of 2016. The urbanization rates of Aba, Ganzi, and Liangshan are relatively low, at 37.86%, 29.26%, and 33.04%, respectively. GDP and the number of vehicles in Liangshan and Panzhihua are obviously higher than in Aba and Ganzi, which means there are more man-made air pollutants in the economic zone (Liangshan and Panzhihua).

Table 1. Resident population, urbanization rate, population density, gross domestic product (GDP), possession of civil motor vehicles and area in the ecological and economic zones of Western Sichuan Plateau.

Region Category	Administrative Area (km ²)	Resident Population (10,000)	Urbanization Rate (%)	GDP (Billion Yuan)	Possession of Civil Motor Vehicles (10,000)	
Aba	Ecological zone	83,016	93.46	37.86	28.13	10.5
Ganzi	Ecological zone	149,599	118.05	29.26	22.98	8.3
Liangshan	Economic zone	60,294	482.22	33.04	140.39	24.9
Panzhihua	Economic zone	7401	123.56	65.34	101.47	15.1

3. Results and Discussion

3.1. Temporal Variations of SO₂, NO₂, and O₃

The variations of SO₂ and NO₂ from 2008 to 2018 and O₃ from 2015 to 2018 in the ecological and economic zones of Western Sichuan Plateau based on monthly average mass concentration are shown in Figure 2. The variations of SO₂ and NO₂ were divided into two periods before and after 2012. SO₂ and NO₂ in Aba showed a downward trend before and after 2012, and the downward trend was more prominent after 2012. However, SO₂ and NO₂ in Ganzi increased before 2012 and decreased after 2012.

O₃ in the ecological zone of the Western Sichuan Plateau, Aba and Ganzi, showed a significant upward trend, with a rising rate of 0.303 $\mu\text{g}\cdot\text{m}^{-3}/\text{month}$ and 0.154 $\mu\text{g}\cdot\text{m}^{-3}/\text{month}$, respectively. Ma et al. [28] reported an increasing trend of surface O₃ in a rural site north of eastern China and indicated that meteorological factors had little influence on long-term O₃ change, which was completely related to emissions. SO₂ and NO₂ in Liangshan showed a trend of first rising and then falling before and after 2012, and the decline of SO₂ was particularly significant after 2012. SO₂ in Panzhihua increased significantly before 2012 and decreased after 2012, while NO₂ showed a slight decline and increase trend before and after 2012. The declining trend of O₃ in Liangshan was very prominent, with a declining rate of 0.36 $\mu\text{g}\cdot\text{m}^{-3}/\text{month}$, while the change range of O₃ in Panzhihua, with the highest urbanization rate, was not obvious. In general, the changes of SO₂ and NO₂ in the ecological zone of Western Sichuan Plateau were weak, while the decrease of SO₂ in the economic zone was very significant and the variation trend of O₃ in the ecological and economic zones was opposite.

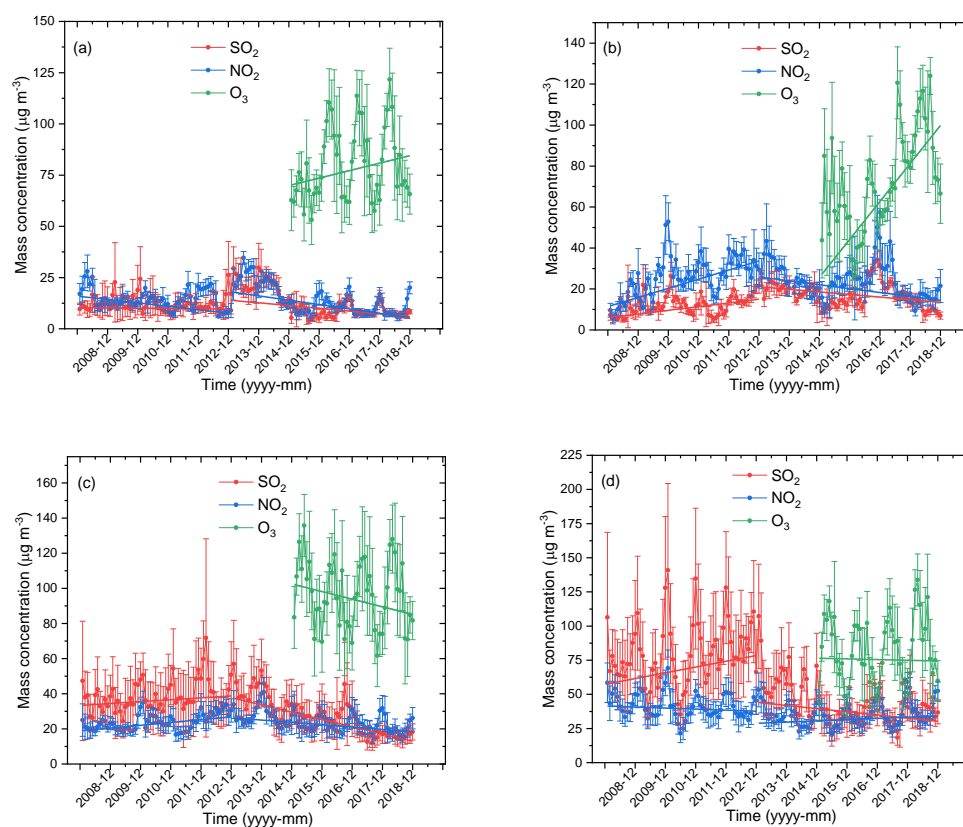


Figure 2. Variations of monthly mean mass concentrations of SO₂, NO₂, and O₃ in (a) Aba, (b) Ganzi, (c) Liangshan, and (d) Panzhihua from 2008 to 2018.

For further analysis, Figure 3 shows the annual variation trend of SO₂, NO₂, and O₃ in the ecological and economic zones of Western Sichuan Plateau, based on the annual average mass concentrations of SO₂ and NO₂ from 2008 to 2018, and O₃ from 2015 to 2018. The concentrations of SO₂ and NO₂ in the ecological area were relatively lower due to the small influence of human activity. The annual variation characteristics of SO₂ and NO₂ in Aba were very similar; their annual concentrations remained stable at about 12 $\mu\text{g}\cdot\text{m}^{-3}$ from 2008 to 2012. From 2012 to 2013, the concentration of SO₂ and NO₂ increased significantly, from, respectively, $12.73 \pm 8.28 \mu\text{g}\cdot\text{m}^{-3}$ to $22.68 \pm 9.25 \mu\text{g}\cdot\text{m}^{-3}$, and from $14.51 \pm 6.92 \mu\text{g}\cdot\text{m}^{-3}$ to $24.12 \pm 7.97 \mu\text{g}\cdot\text{m}^{-3}$, then decreased to $7.79 \pm 4.96 \mu\text{g}\cdot\text{m}^{-3}$ and $10.27 \pm 4.88 \mu\text{g}\cdot\text{m}^{-3}$ in 2015, and then stabilized to 2018. The average O₃ concentration in Aba increased from $66.56 \pm 14.26 \mu\text{g}\cdot\text{m}^{-3}$ to $86.37 \pm 23.17 \mu\text{g}\cdot\text{m}^{-3}$ from 2015 to 2018. The annual average concentration of NO₂ in Ganzi was always higher than that of SO₂ from 2008 to 2018, but the annual variations of SO₂ and NO₂ were relatively close. SO₂ wavelike variation went up from 2008 to 2013, then went down, then rose again from 2015 to 2016,

then fell again; the lowest concentration was $9.05 \pm 7.81 \mu\text{g}\cdot\text{m}^{-3}$ in 2008, and the highest concentration was $22.62 \pm 9.74 \mu\text{g}\cdot\text{m}^{-3}$ in 2016. The lowest concentration of NO_2 in Ganzi was $15.77 \pm 8.72 \mu\text{g}\cdot\text{m}^{-3}$ in 2008, and the highest concentration was $32.83 \pm 8.96 \mu\text{g}\cdot\text{m}^{-3}$ in 2012. Overall, SO_2 and NO_2 in Ganzi showed an upward and then a downward trend, and the annual average concentrations of 2008 and 2018 were relatively close. The average O_3 concentration in Ganzi dropped from $62.47 \pm 26.75 \mu\text{g}\cdot\text{m}^{-3}$ in 2015 to $52.64 \pm 19.97 \mu\text{g}\cdot\text{m}^{-3}$ in 2016, and then rose to $95.45 \pm 22.86 \mu\text{g}\cdot\text{m}^{-3}$ in 2018.

The biggest difference between annual variation characteristics of SO_2 and NO_2 in the economic and ecological zones was that the annual average concentration of SO_2 was almost always higher than that of NO_2 . In 2008–2016, the mean SO_2 concentration in Liangshan was always higher than that of NO_2 . The mean SO_2 concentration increased from $35.84 \pm 16.76 \mu\text{g}\cdot\text{m}^{-3}$ in 2008 to $43.41 \pm 16.72 \mu\text{g}\cdot\text{m}^{-3}$ in 2013 and then fell irregularly to $16.44 \pm 4.71 \mu\text{g}\cdot\text{m}^{-3}$ in 2018. In general, SO_2 in Liangshan showed a downward trend, and the concentration in 2018 was 54.13% lower than that in 2008. NO_2 wavelike variation in Liangshan increased from $21.82 \pm 5.15 \mu\text{g}\cdot\text{m}^{-3}$ in 2008 to $29.3 \pm 5.95 \mu\text{g}\cdot\text{m}^{-3}$ in 2012, (34.4%), and then fell to $20.52 \pm 6.62 \mu\text{g}\cdot\text{m}^{-3}$ in 2018. The annual average concentration of O_3 in Liangshan showed a similar trend to Ganzi, first decreasing from $99.93 \pm 27.66 \mu\text{g}\cdot\text{m}^{-3}$ in 2015 to $92.49 \pm 27.47 \mu\text{g}\cdot\text{m}^{-3}$ in 2016, and then rising to $98.68 \pm 28.85 \mu\text{g}\cdot\text{m}^{-3}$ in 2018. Panzhihua has the highest urbanization among the four regions and the largest change range of SO_2 annual average concentration. The SO_2 concentration in Panzhihua was always higher than that of NO_2 in 2008–2018. The average concentration of SO_2 increased from $76.08 \pm 32.77 \mu\text{g}\cdot\text{m}^{-3}$ in 2008 to $86.52 \pm 33.16 \mu\text{g}\cdot\text{m}^{-3}$ in 2012, then fell sharply to $33.55 \pm 14.65 \mu\text{g}\cdot\text{m}^{-3}$ in 2015, then increased slightly to about $39 \mu\text{g}\cdot\text{m}^{-3}$ in 2018. The average concentration of NO_2 in Panzhihua decreased from $44.47 \pm 12.01 \mu\text{g}\cdot\text{m}^{-3}$ in 2008 to $31.82 \pm 9.67 \mu\text{g}\cdot\text{m}^{-3}$ in 2015, (28.45%), and then went up to $38.57 \pm 10.49 \mu\text{g}\cdot\text{m}^{-3}$ in 2018. The annual variation of O_3 in Panzhihua showed a trend of first decreasing and then increasing, from $90.34 \pm 22.89 \mu\text{g}\cdot\text{m}^{-3}$ in 2014 to $75.79 \pm 26.38 \mu\text{g}\cdot\text{m}^{-3}$ in 2016, and then recovering to $93.05 \pm 33.49 \mu\text{g}\cdot\text{m}^{-3}$ in 2018.

In general, the annual variation of gas pollutants in the Western Sichuan Plateau, a relatively clean area, was different from that in large and medium-sized cities. The overall variations of SO_2 and NO_2 in the Western Sichuan Plateau were not very large, showing a trend of first increasing and then decreasing.

Overall, SO_2 and NO_2 in Aba, Ganzi, and Liangshan showed a small change range, and both showed a trend of first increasing and then decreasing in 2008–2018. However, after China's 11th Five-Year Plan (2006–2010), SO_2 concentration in large and medium-sized cities [14,29,30] showed an obvious downward trend, while NO_2 showed a fluctuating or rising trend. The significant decrease of SO_2 concentration in Panzhihua from 2012 to 2015 was also related to the emission reduction measures in China's 12th Five-Year Plan (2011–2015). This shows that after the 11th Five-Year Plan, the emission reduction requirements for SO_2 and NO_2 proposed by the government had no obvious impact on the relatively clean Western Sichuan Plateau. The annual variations of O_3 in the ecological and economic zones of the Western Sichuan Plateau were like those in Eastern China. Li et al. [20] showed that O_3 increased significantly during the period of 2013–2017 in the North China Plain, the Pearl River Delta, Chengdu-Chongqing urban agglomeration, and Southeastern China, while the O_3 pollution in parts of the northwest, southwest, and northeast of the country was relatively light.

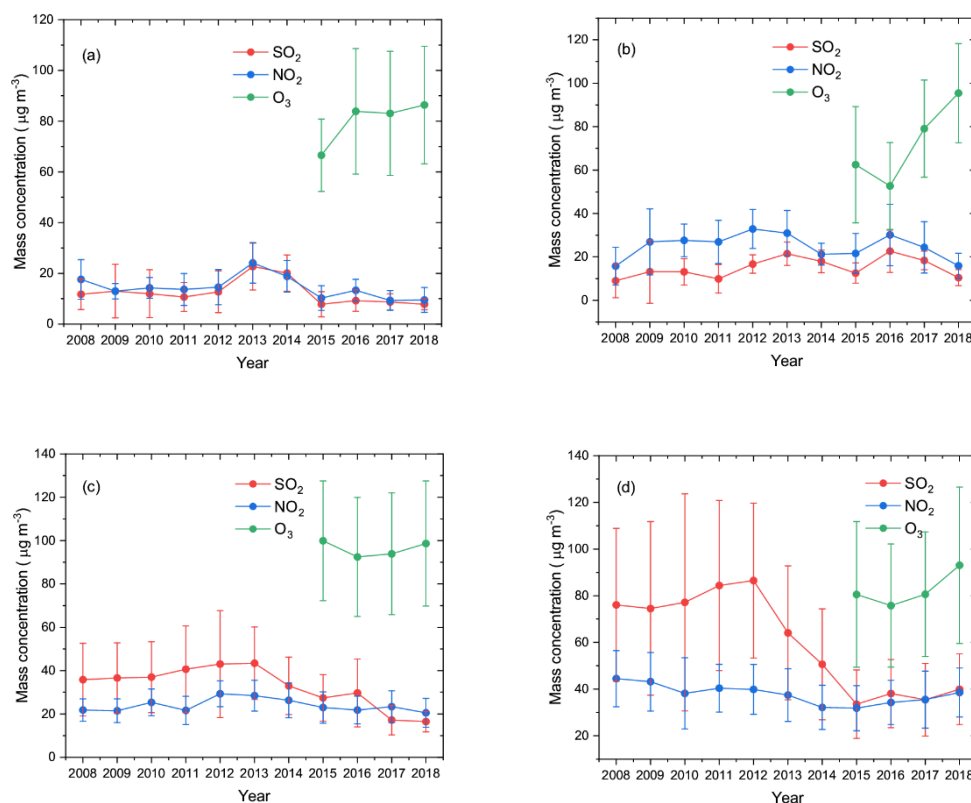


Figure 3. Variations of annual mean mass concentrations of SO₂, NO₂, and O₃ in (a) Aba, (b) Ganzi, (c) Liangshan, and (d) Panzhihua from 2008 to 2018.

Figure 4 shows the annual variation characteristics of the average concentrations of SO₂, NO₂, and O₃ in different seasons in the ecological and economic zones of Western Sichuan Plateau. Figure 4a–c show the annual variations of SO₂, NO₂, and O₃ in different seasons in Aba. The annual changes of SO₂ in spring and winter were more significant, rising first and then falling, reaching the maximum value in 2013. NO₂ increased first and then decreased in all seasons, with the largest change in summer, reaching a maximum of 30.33 µg·m⁻³ in 2013. In both summer and winter, O₃ showed an upward and then a downward trend, while in spring, it showed a very significant upward trend, with an increase of about 55.4% from 2015 to 2018. Figure 4d–f show the annual variations of seasonal average concentrations of SO₂, NO₂, and O₃ in Ganzi. In the autumn, SO₂ showed the most significant annual change, rising from 9.4 µg·m⁻³ in 2008 to 31.5 µg·m⁻³ in 2016, and finally falling to 9.3 µg·m⁻³ in 2018. NO₂ fluctuated in all seasons, with the largest change in spring, increasing about 233.6% from 2008 to 2013, and decreasing about 58.1% from 2013 to 2018. In spring and autumn, O₃ showed a downward and then an upward trend, while in summer, it showed a significant upward trend, with an increase of about 88.8% from 2015 to 2018. Both SO₂ (Figure 4g) and NO₂ (Figure 4h) in Liangshan showed the most obvious annual change range in winter; SO₂ and NO₂ reached the maximum value in 2011 and 2013, respectively. In spring, summer, and autumn, O₃ in Liangshan (Figure 4i) showed a trend of first declining and then rising, while in winter it showed a trend of first rising and then falling, but the change range was small, decreasing by 5.8 µg·m⁻³ in 2017–2018. The seasonal average concentration of SO₂ in Panzhihua (Figure 4j) showed the largest change in winter, with a decrease of 69.9% from 121.9 µg·m⁻³ in 2009 to 36.6 µg·m⁻³ in 2018. NO₂ in Panzhihua (Figure 4k) showed a trend of first declining and then rising in all four seasons; the change ranges of spring and winter were large, and the seasonal average concentration in spring and winter reached the minimum value in 2016 and 2015, respectively. O₃ in Panzhihua (Figure 4l) showed a trend of first declining and then rising in all four seasons, with a large change range in spring and summer, and an increase of about 26% in spring from 2016 to 2018.

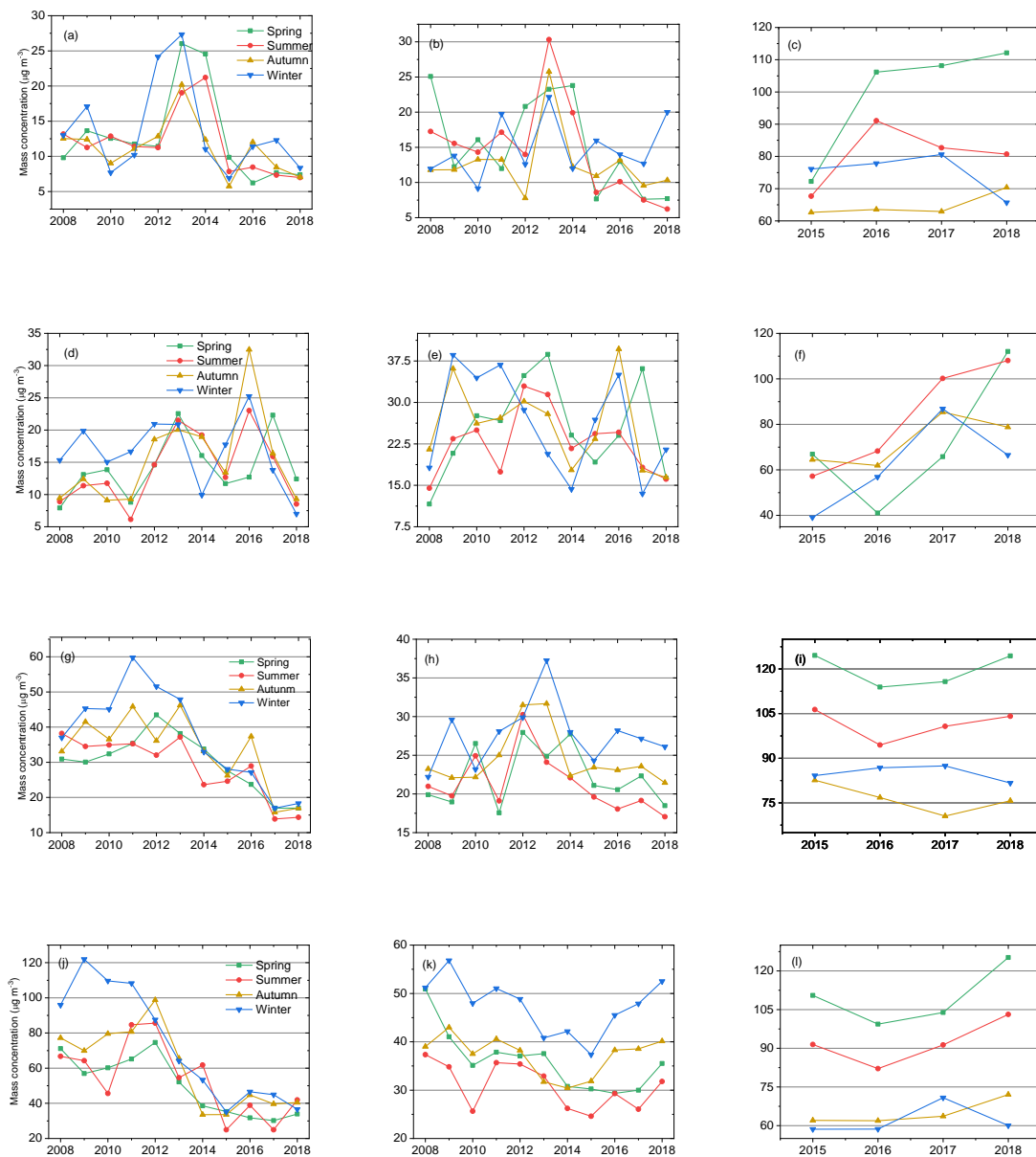


Figure 4. Annual variations of three air pollutants in different seasons: (a) SO₂, (b) NO₂, and (c) O₃ in Aba; (d) SO₂, (e) NO₂, and (f) O₃ in Ganzi; (g) SO₂, (h) NO₂, and (i) O₃ in Liangshan; (j) SO₂, (k) NO₂, and (l) O₃ in Panzhihua.

The average mass concentration ratio of SO₂ and NO₂ (SO₂/NO₂) can represent the energy structure characteristics of a local region [31]. For example, in regions with more developed industries, a large amount of sulfur-containing fuel is consumed and SO₂ is emitted, thus, SO₂/NO₂ is larger. If an area has a mass of cars and emits more NO_x, the ratio is relatively small. In the 1990s, SO₂/NO₂ in North America and some eastern and central European countries exceeded 1, indicating that these regions had developed industries and the emissions were mainly SO₂ [31]. Figure 5 shows the change characteristics of SO₂/NO₂ based on monthly average concentration in the ecological and economic zones of Western Sichuan Plateau from 2008 to 2018. As can be seen, SO₂/NO₂ of Aba and Ganzi in the ecological zone showed an upward trend, while SO₂/NO₂ of Liangshan and Panzhihua in the economic zone showed a downward trend. From 2008 to 2018, SO₂/NO₂ of Aba fluctuated and showed a slight upward trend, with an average increase of 8.9×10^{-5} /month. Overall, SO₂/NO₂ of Ganzi was smaller than that of Aba, and the overall SO₂/NO₂ fluctuated and had a more obvious upward trend,

with an average increase of 0.002/month, and SO_2/NO_2 stayed below 1.0 most of the time. SO_2/NO_2 in both Liangshan and Panzhihua showed an obvious downward trend, with an average decline of $-0.007/\text{month}$ in Liangshan and a slightly higher rate in Panzhihua, $-0.008/\text{month}$. According to the annual average concentration ratio of SO_2 and NO_2 (not shown), before 2017, the annual SO_2/NO_2 in Liangshan was greater than 1.0, and the maximum value (about 1.88) appeared in 2011, while in 2017 and 2018, SO_2/NO_2 was less than 1.0 (0.73 and 0.80, respectively). The annual SO_2/NO_2 in Panzhihua was higher than that in Liangshan; from 2008 to 2012, it showed a quasi-linear growth trend, increasing from 1.71 in 2008 to 2.17 in 2012, and then significantly decreasing, reaching the lowest value of about 0.99 in 2017.

The rising trend of SO_2/NO_2 in the Western Sichuan Plateau ecological area and the declining trend in the economic area indicates that the energy consumption structure of these two areas is quite different. The rising trend of the ecological area indicates that industrial SO_2 emission of in the area have increased in the past 11 years compared with NO_x emissions from transportation. However, the declining trend of SO_2/NO_2 in the economic zone in the past decade is similar to that of some large and medium-sized cities in eastern China, indicating that the SO_2 emission reduction effect in this region was relatively obvious, while the NO_x emission reduction effect was not significant. It is worth noting that China's 11th Five-Year Plan (2011–2015) put forward specific SO_2 and NO_x emission reduction targets, since SO_2/NO_2 in most Chinese cities showed a declining trend, and SO_2/NO_2 in the ecological zone of Western Sichuan Plateau, which is cleaner than the large and medium-sized cities, showed an increasing trend. This suggests that the SO_2 pollution level in clean areas is relatively light, but its growth trend cannot be ignored.

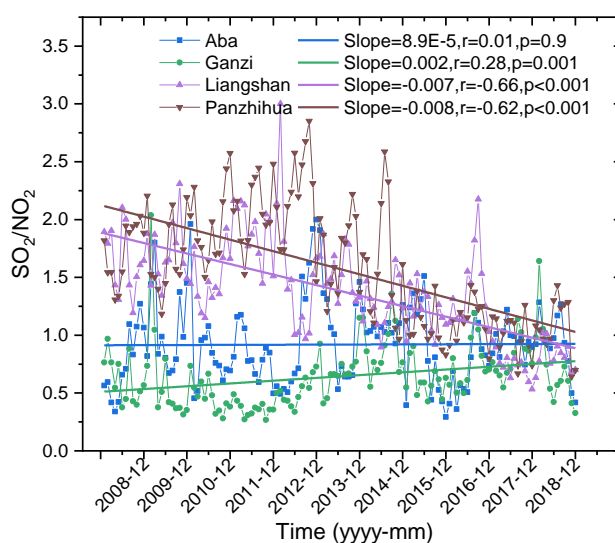


Figure 5. Variations of concentration ratio of SO_2 to NO_2 in Aba, Ganzi, Liangshan, and Panzhihua from 2008 to 2018.

Due to the lack of emissions data of SO_2 and NO_x in the ecological and economic zones of Western Sichuan Plateau, the energy consumption of unit added industrial value and the passenger and freight turnover of highways were selected to indirectly characterize the emission change characteristics of SO_2 and NO_x , respectively. As can be seen from Figure 6a, from 2008 to 2016, the energy consumption of unit added industrial value in Aba first increased and then decreased, while other regions showed a significant downward trend, indicating that industrial consumption of standard coal equivalent (SCE) in other regions except Aba declined year by year. Compared with the change characteristics of SO_2 concentration from 2008 to 2018 (Figure 3), it was found that the variations of SO_2 are not consistent with industrial emissions, indicating that in these relatively clean areas, other sources besides industrial emissions will also contribute significantly to SO_2 ; for instance, biomass combustion in Qinghai Plateau [32] makes an important contribution to SO_2 . The passenger and freight turnover of

highways (Figure 6b) shows an overall upward trend, first increasing and then decreasing before and after 2013, which is close to the changing trend of NO_2 concentration (Figure 3), indicating that motor vehicle emissions made a prominent contribution to the change in NO_2 concentration.

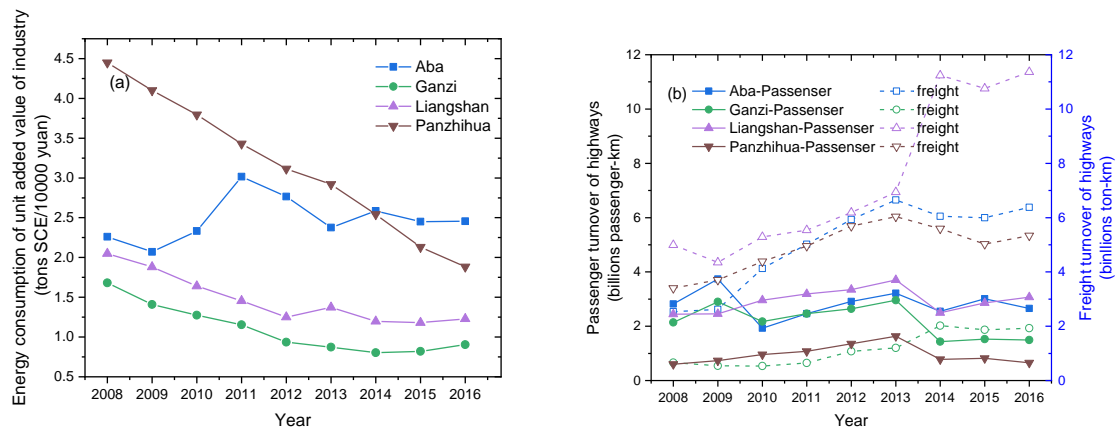


Figure 6. (a) Energy consumption per unit industrial added value and (b) highway passenger and freight turnover in Aba, Ganzi, Liangshan, and Panzhihua in 2008–2016.

3.2. Effects of Meteorology on SO_2 , NO_2 and O_3

Emissions and meteorological conditions are predominant in the air pollutant trend; emissions exceeding atmospheric environmental capacity is the basic cause of air pollution [33,34]. In order to analyze the influence of meteorological factors on gaseous air pollutants in the ecological and economic zones of Western Sichuan Plateau, the relationships between SO_2 , NO_2 , O_3 , and routine meteorological parameters (mean monthly average temperature, maximum and minimum monthly average temperature, mean relative humidity, monthly total precipitation, monthly sunshine duration, mean wind speed, and maximum wind speed) were discussed by using monthly mean meteorological data from 2008 to 2017. Figure 7 shows these relationships in Aba, in the ecological zone. SO_2 and NO_2 concentrations were less dependent on meteorological parameters in the ecological zone with less anthropogenic emission. The relationship between O_3 and meteorological parameters in Figure 7 implies that a high concentration of O_3 was more likely to form under the meteorological conditions of high temperature, low relative humidity, long sunshine duration, and especially high wind speed.

The relationships of SO_2 , NO_2 , O_3 , and meteorological parameters in Panzhihua of the economic zone of Western Sichuan Plateau are shown in Figure 8. In the economic zone, with more man-made emissions than the ecological zone, gaseous pollutants are more significantly affected by meteorological parameters with a higher coefficient of determination. SO_2 and NO_2 decreased with increased temperature, humidity, precipitation, and wind speed. O_3 was positively correlated with temperature, sunshine duration, and wind speed and negatively correlated with relative humidity. The dependence of gaseous pollutants on meteorological factors was more prominent in the economic zone than in the ecological zone, which may have to do with local emissions and special topography [21].

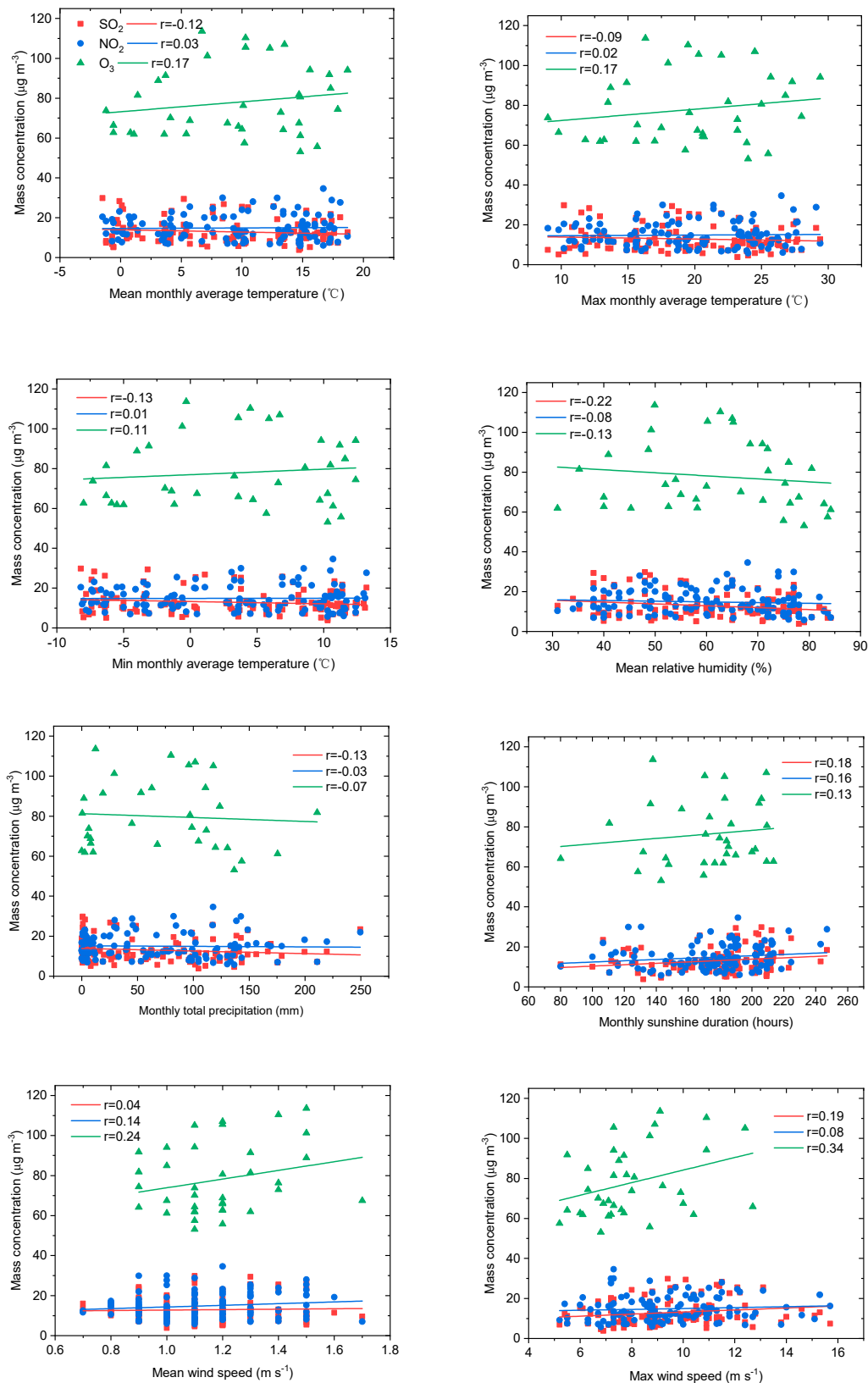


Figure 7. Relationships between SO₂, NO₂, O₃ and routine meteorological parameters (mean monthly average temperature, maximum and minimum monthly average temperature, mean relative humidity, monthly total precipitation, monthly sunshine duration, mean wind speed, and maximum wind speed) in Aba of the ecological zone of Western Sichuan Plateau.

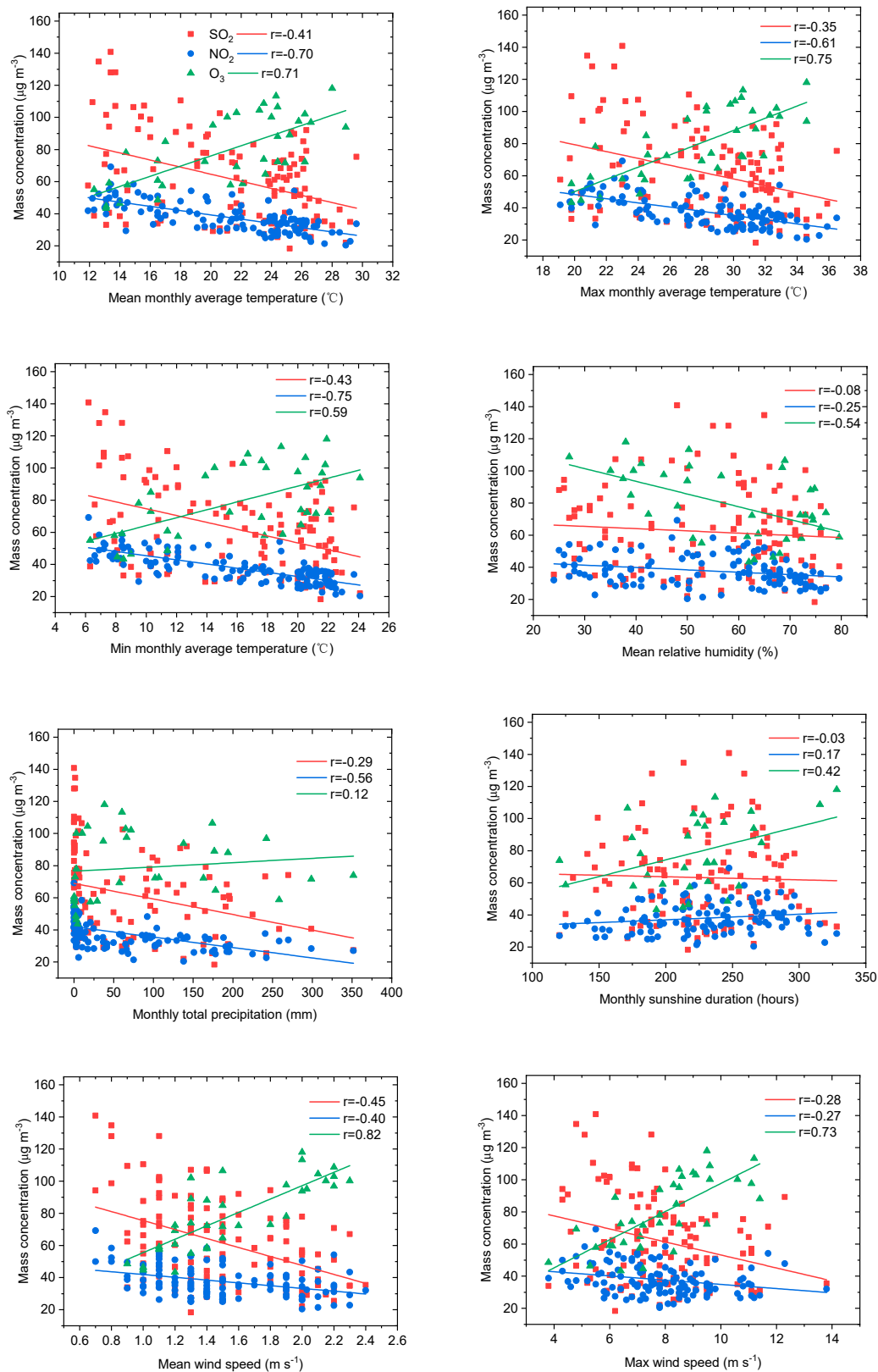


Figure 8. Relationships between SO₂, NO₂, O₃, and routine meteorological parameters (mean monthly average temperature, maximum and minimum monthly average temperature, mean relative humidity, monthly total precipitation, monthly sunshine duration, mean wind speed, and maximum wind speed) in Panzhuhua of the economic zone of Western Sichuan Plateau.

3.3. Characteristics of SO₂, NO₂, and O₃

Figure 9 shows the multi-year average mass concentrations of SO₂, NO₂, and O₃ in the Western Sichuan Plateau ecological and economic zones. The average mass concentrations of O₃ in Aba, Ganzi, Liangshan, and Panzhihua were significantly higher than that of SO₂ and NO₂. Liangshan suffered the most serious O₃ pollution of the four regions, with a mean mass concentration of $96.25 \pm 28.17 \mu\text{g}\cdot\text{m}^{-3}$, followed by Panzhihua, $83.96 \pm 29.17 \mu\text{g}\cdot\text{m}^{-3}$. O₃ pollution in the ecological region, at higher altitude, was lighter than in the economic zone, with mean mass concentrations of $79.99 \pm 23.41 \mu\text{g}\cdot\text{m}^{-3}$ and $72.61 \pm 28.28 \mu\text{g}\cdot\text{m}^{-3}$, respectively, in Aba and Ganzi. The mean mass concentrations of SO₂ and NO₂ in the ecological zone were lower than in the economic zone. The mean mass concentration of SO₂ in Aba and Ganzi was similar, at $12.39 \pm 8.31 \mu\text{g}\cdot\text{m}^{-3}$ and $15.02 \pm 8.51 \mu\text{g}\cdot\text{m}^{-3}$, respectively, and in Panzhihua, it was significantly higher than in Liangshan, at $60.05 \pm 35.25 \mu\text{g}\cdot\text{m}^{-3}$ and $32.73 \pm 18.02 \mu\text{g}\cdot\text{m}^{-3}$, respectively. Among the four regions, Panzhihua has the most serious SO₂ pollution, and Aba has the least, the multi-year average concentration of SO₂ in Panzhihua is about five times that in Aba. For NO₂, the average mass concentration was highest in Panzhihua, at $37.81 \pm 11.98 \mu\text{g}\cdot\text{m}^{-3}$ and lowest in Aba, at $14.37 \pm 7.07 \mu\text{g}\cdot\text{m}^{-3}$. The level of NO₂ pollution in Ganzi and Liangshan was moderate, and the average concentration of NO₂ was similar, $24.99 \pm 11.59 \mu\text{g}\cdot\text{m}^{-3}$ and $23.9 \pm 7.17 \mu\text{g}\cdot\text{m}^{-3}$, respectively. Zhao et al. [21] analyzed the characteristics of six criteria for air pollutants in the Sichuan basin in 2015–2017, and showed that the average SO₂ concentration in Chengdu, the provincial city, was close to that in the ecological zone due to effective desulfurization measures and strict SO₂ emission reduction requirements, while the average NO₂ and O₃ concentration in Chengdu was significantly higher than that in the ecological and economic zones in this study.

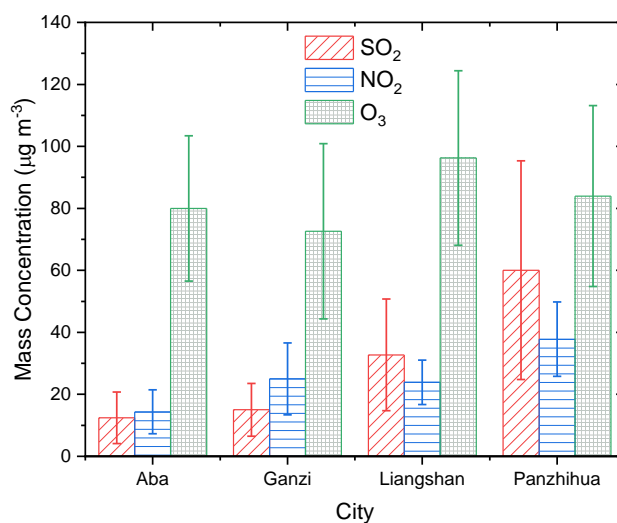


Figure 9. Multi-year mean mass concentrations of SO₂, NO₂, and O₃ in Aba, Ganzi, Liangshan, and Panzhihua.

Figure 10 shows the seasonal average mass concentrations of SO₂, NO₂, and O₃ in the ecological and economic zones of Western Sichuan Plateau. The seasonal variation of O₃ is more notable than that of SO₂ or NO₂. The seasonal distribution of O₃ in Aba and Liangshan is similar, with the highest concentration in spring and the lowest in autumn, presenting a seasonal distribution as spring > summer > winter > autumn. The average concentration of O₃ in Aba was $99.8 \pm 21.54 \mu\text{g}\cdot\text{m}^{-3}$ and $64.83 \pm 12.95 \mu\text{g}\cdot\text{m}^{-3}$, and that in Liangshan was $119.64 \pm 22.47 \mu\text{g}\cdot\text{m}^{-3}$ and $76.42 \pm 20.63 \mu\text{g}\cdot\text{m}^{-3}$ in spring and autumn, respectively. The seasonal distribution of O₃ in Ganzi was summer > autumn > spring > winter, with an average concentration of $83.67 \pm 31.85 \mu\text{g}\cdot\text{m}^{-3}$ in summer and $64.94 \pm 24.14 \mu\text{g}\cdot\text{m}^{-3}$ in winter. The concentration of O₃ in Panzhihua reached its highest in spring $107.64 \pm 19.97 \mu\text{g}\cdot\text{m}^{-3}$, and its lowest in winter, $64.61 \pm 19.2 \mu\text{g}\cdot\text{m}^{-3}$, indicating a seasonal distribution of spring > summer > autumn > winter.

The average concentration of SO₂ in Ganzi and Liangshan was the same, with a seasonal distribution of winter > autumn > spring > summer. The average concentration was $17.4 \pm 9.7 \mu\text{g}\cdot\text{m}^{-3}$ and $13.94 \pm 7.28 \mu\text{g}\cdot\text{m}^{-3}$ in Ganzi and was $38.46 \pm 21.88 \mu\text{g}\cdot\text{m}^{-3}$ and $28.87 \pm 15.15 \mu\text{g}\cdot\text{m}^{-3}$ in Liangshan in winter and summer, respectively. The maximum concentration of SO₂ in Aba occurred in winter, while the minimum concentration occurred in autumn, at $13.84 \pm 9.74 \mu\text{g}\cdot\text{m}^{-3}$ and $11.24 \pm 7.38 \mu\text{g}\cdot\text{m}^{-3}$; the seasonal distribution was winter > spring > summer > autumn. The mean concentration of SO₂ in Panzhihua was the highest in winter, $75.44 \pm 45.67 \mu\text{g}\cdot\text{m}^{-3}$, and the lowest in spring $49.99 \pm 25.32 \mu\text{g}\cdot\text{m}^{-3}$.

Ganzi, Liangshan, and Panzhihua had the same NO₂ seasonal distribution of winter > autumn > spring > summer. The difference in NO₂ mean concentration in Ganzi and Liangshan between the four seasons was not significant; the maximum and minimum concentrations in Ganzi were $27.16 \pm 13.5 \mu\text{g}\cdot\text{m}^{-3}$ and $22.66 \pm 8.68 \mu\text{g}\cdot\text{m}^{-3}$, respectively, and the difference between winter and summer was about $4.5 \mu\text{g}\cdot\text{m}^{-3}$. The maximum and minimum NO₂ mean concentrations in Liangshan were $27.72 \pm 8.93 \mu\text{g}\cdot\text{m}^{-3}$ and $21.37 \pm 5.31 \mu\text{g}\cdot\text{m}^{-3}$, respectively, and the difference in concentration between winter and summer was about $6.3 \mu\text{g}\cdot\text{m}^{-3}$. The average concentration of NO₂ in Panzhihua in the four seasons was significantly different; the maximum concentration was $47.14 \pm 11.88 \mu\text{g}\cdot\text{m}^{-3}$ in winter, the minimum concentration was $30.89 \pm 8.13 \mu\text{g}\cdot\text{m}^{-3}$ in summer, and the difference was about $16.2 \mu\text{g}\cdot\text{m}^{-3}$. The seasonal distribution of NO₂ in Aba was different from the other regions, with an average concentration of $15.39 \pm 7.57 \mu\text{g}\cdot\text{m}^{-3}$ in spring and the lowest concentration of $12.71 \pm 6.19 \mu\text{g}\cdot\text{m}^{-3}$ in autumn.

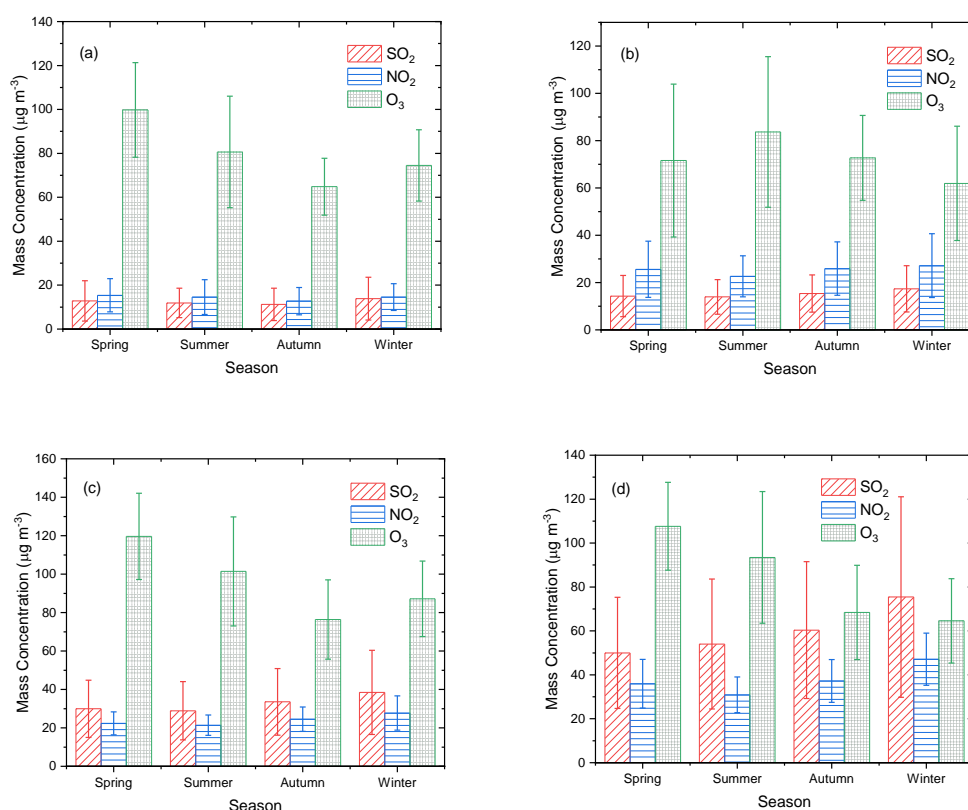


Figure 10. Seasonal mean mass concentrations of SO₂, NO₂, and O₃ in (a) Aba, (b) Ganzi, (c) Liangshan, and (d) Panzhihua.

Figure 11 shows the monthly distribution characteristics of the three pollutants. High concentrations of O₃ in Ganzi appeared in July, August, and September, and in Aba, Liangshan, and Panzhihua in March, April, and May. The maximum O₃ concentration in Ganzi was $94.63 \pm 29.89 \mu\text{g}\cdot\text{m}^{-3}$ in August, and the minimum was $56.05 \pm 22.94 \mu\text{g}\cdot\text{m}^{-3}$ in January. The maximum and

minimum concentrations of O_3 in Aba occurred in April and September, respectively, at $103.46 \pm 22.15 \mu\text{g}\cdot\text{m}^{-3}$ and $62.16 \pm 16.47 \mu\text{g}\cdot\text{m}^{-3}$. The maximum concentration of O_3 in Liangshan occurred in May at $123.40 \pm 25.3 \mu\text{g}\cdot\text{m}^{-3}$ and the minimum concentration of $72.49 \pm 21.66 \mu\text{g}\cdot\text{m}^{-3}$ in September. The maximum and minimum concentrations of O_3 in Panzhuhua occurred in April and December, at $109.65 \pm 20.66 \mu\text{g}\cdot\text{m}^{-3}$ and $56.17 \pm 16.49 \mu\text{g}\cdot\text{m}^{-3}$, respectively. The differences between maximum and minimum monthly concentrations in the economic zone were significantly higher than in the ecological zone, among which the difference in Panzhuhua was the largest, about $53.48 \mu\text{g}\cdot\text{m}^{-3}$, and the difference in Ganzi was the smallest, about $38.5 \mu\text{g}\cdot\text{m}^{-3}$, indicating that the monthly change of O_3 with a longer residence time was more significant in the area with more artificial emissions than in the clean area with less artificial emissions.

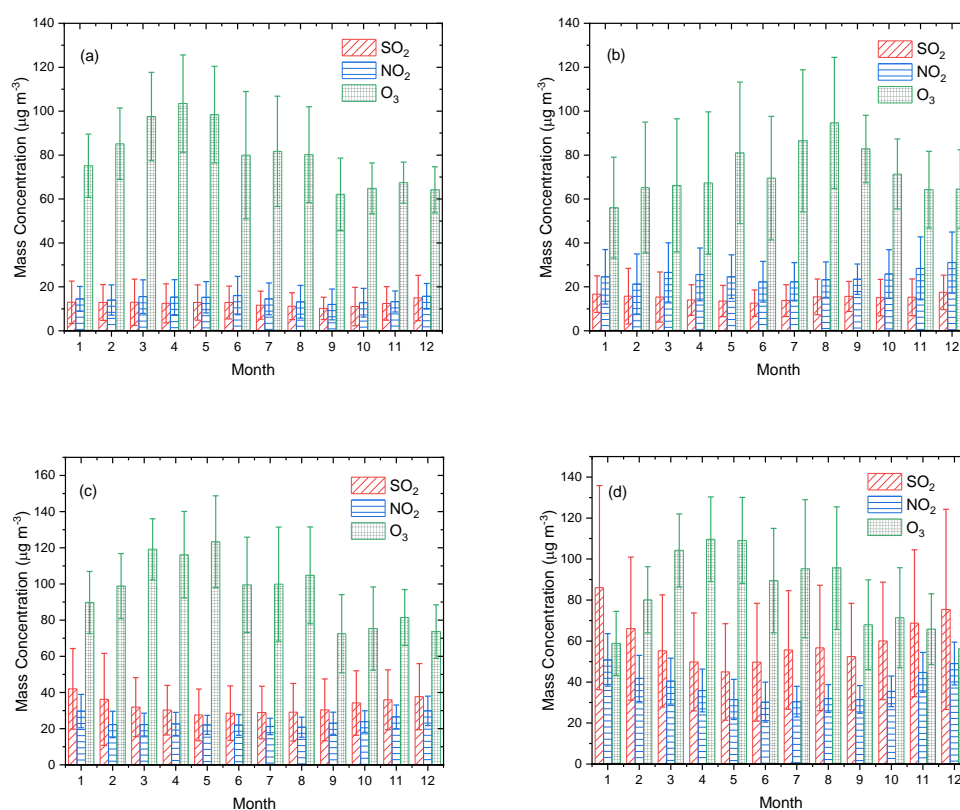


Figure 11. Monthly mean mass concentrations of SO_2 , NO_2 , and O_3 in (a) Aba, (b) Ganzi, (c) Liangshan, and (d) Panzhuhua.

The monthly mean concentrations of SO_2 in the ecological zone did not change significantly. The maximum and minimum concentrations in Aba appeared in December and September, respectively, and the difference was about $4.76 \mu\text{g}\cdot\text{m}^{-3}$, while the maximum and minimum concentrations in Ganzi appeared in December and June, respectively, and the difference was about $4.9 \mu\text{g}\cdot\text{m}^{-3}$. Artificial emissions were more prominent in the economic zone, and the monthly variations of average concentrations of SO_2 were more obvious. The maximum and minimum SO_2 concentration in Liangshan occurred in January and May, with a difference of $14.43 \mu\text{g}\cdot\text{m}^{-3}$, and in Panzhuhua occurred in January and May, respectively, at $86.01 \pm 49.78 \mu\text{g}\cdot\text{m}^{-3}$ and $44.96 \pm 23.54 \mu\text{g}\cdot\text{m}^{-3}$, with a difference of $41.05 \mu\text{g}\cdot\text{m}^{-3}$. The range of variation of monthly average NO_2 concentration in Aba was the lowest in the ecological and economic zones; the maximum and minimum concentrations appeared in June and September, with a difference of $6.05 \mu\text{g}\cdot\text{m}^{-3}$, and the maximum and minimum concentrations in Ganzi appeared in December and February, with a difference of $9.82 \mu\text{g}\cdot\text{m}^{-3}$, respectively. The monthly average NO_2 concentration in Panzhuhua showed the most prominent variation characteristics, with a mean maximum of $50.82 \pm 12.79 \mu\text{g}\cdot\text{m}^{-3}$ in January, minimum of $30.21 \pm 9.79 \mu\text{g}\cdot\text{m}^{-3}$ in June, and a

difference of $20.61 \mu\text{g}\cdot\text{m}^{-3}$. The maximum and minimum monthly average concentrations of NO_2 in Liangshan occurred in December and August, respectively, with a difference of $9.01 \mu\text{g}\cdot\text{m}^{-3}$.

3.4. Correlation between SO_2 , NO_2 , and O_3

It is generally believed that CO and NO_x pollutants are discharged by mobile sources, while SO_2 and NO_x pollutants are discharged by point sources [35]. Since mobile sources do not discharge a large amount of SO_2 and point sources produce both SO_2 and NO_2 , if the correlation between SO_2 and NO_2 is high, the point source is more prominent. On the other hand, if the correlation is low, the mobile source is more prominent. Figure 12 shows the correlation between SO_2 and NO_2 in Aba, Ganzi, Liangshan, and Panzhihua. SO_2 and NO_2 in the ecological and economic zones show a positive correlation with a significance test of 99% confidence. The correlation between SO_2 and NO_2 in Ganzi is the highest, with a Pearson coefficient $r = 0.55$, followed by Panzhihua, with $r = 0.47$, which indicates that the emission characteristics of SO_2 and NO_2 in these two regions are relatively similar, SO_2 and NO_2 are discharged mainly from point sources and point sources contribute more to local air pollutants than mobile sources. The lowest correlation between SO_2 and NO_2 is found in Aba, with a correlation coefficient of 0.27, while that in Liangshan is slightly higher, with $r = 0.31$, indicating that the contribution of point sources to SO_2 and NO_2 pollution in these two areas is not very prominent. In general, although SO_2 and NO_2 in the ecological and economic zones were responsible less pollution than in other large and medium-sized cities in China, the emission source characteristics of SO_2 and NO_2 in this region were different from those of the other cities. Mao et al. [36] reported a strong positive correlation between SO_2 and NO_2 in Chongqing, Wuhan, and Nanjing, three metropolises along the Yangtze River, indicating the similar origins and elimination processes of SO_2 and NO_2 .

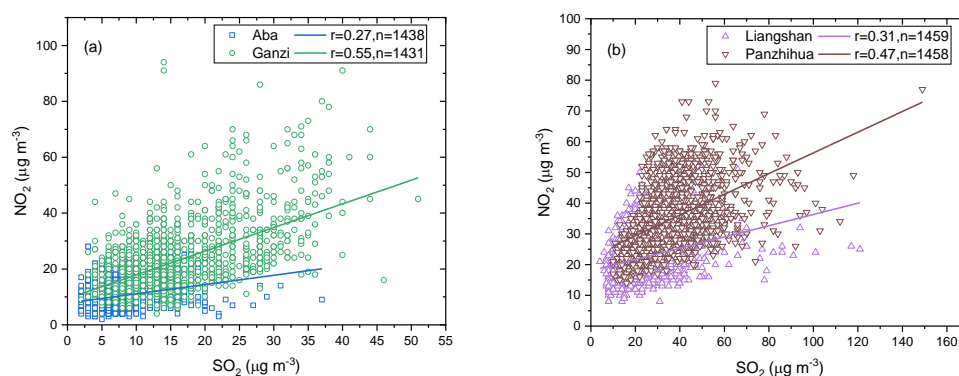


Figure 12. Correlation between SO_2 and NO_2 in the (a) ecological zone and (b) economic zone in 2015–2018.

O_3 is an important secondary gaseous pollutant in the urban atmosphere, and NO_x plays a very important role in its formation as the main precursor. The formation of surface O_3 is highly dependent on solar radiation intensity, the absolute concentration of O_3 and volatile organic compounds (VOCs), and the ratio of O_3 to VOCs, while the local surface concentration of O_3 is affected by meteorological elements, local precursor emissions, and the close and long-distance transport of O_3 and precursor [37–39]. As shown in Figure 13a,b, the correlations between NO_2 and O_3 in the ecological and economic zones of Western Sichuan Plateau were analyzed by using the daily mass concentration from 2015 to 2018. The correlations between NO_2 and O_3 in the four regions were negatively correlated, and all of them passed the significance test of 99% confidence. Among the four regions, Ganzi has the highest Pearson correlation coefficient (-0.34), indicating that this region is where NO_2 contributes the most to O_3 generation as a precursor. Aba, Liangshan, and Panzhihua showed a relatively low correlation between NO_2 and O_3 , with a Pearson correlation coefficient of -0.24 , -0.22 , and -0.26 , respectively. The correlations between NO_2 and O_3 in the metropolitan areas of China, such as the Beijing–Tianjin–Hebei region, Yangtze River Delta, Pearl River Delta, and Chengdu–Chongqing city

cluster [2,36,40] are significantly higher than in the ecological and economic zones of the Western Sichuan Plateau in our study, indicating that NO_2 contributes more to O_3 in large and medium-sized urban areas with high NO_2 emissions. The contribution of local precursors of O_3 can better explain the formation of O_3 , while in regions with less NO_2 emission, O_3 may be more affected by the regional background. de Souza et al. [41] also indicated that when the correlation between NO_2 and O_3 is high, the local precursor plays a leading contributory role, and when the correlation is low, the variation of O_3 concentration is mainly affected by the O_3 concentration in the regional background.

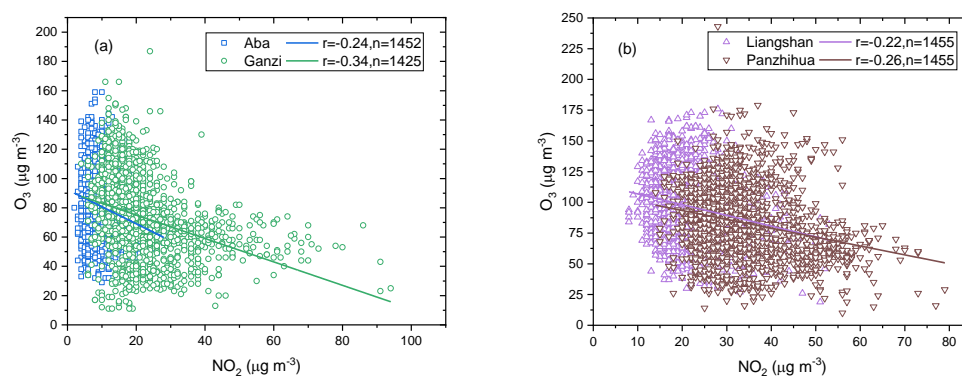


Figure 13. Correlation between NO_2 and O_3 in the (a) ecological zone and (b) economic zone in 2015–2018.

4. Conclusions

Based on daily average mass concentration data of SO_2 and NO_2 from 2008 to 2018 and O_3 from 2015 to 2018 in Aba, Ganzi, Liangshan, and Panzhihua in Western Sichuan Plateau, the characteristics and change trend of the three gas pollutants and their correlations in the relatively clean ecological and economic zones were analyzed. The main conclusions are as follows:

- (1) On the whole, the change ranges of SO_2 and NO_2 in Aba, Ganzi, and Liangshan were not large, and they all showed a trend of first increasing and then decreasing. The change range of SO_2 in Panzhihua was the most obvious, and the decrease range was very significant from 2012 to 2015. During the period 2015–2018, except for Aba, O_3 showed a trend of first declining and then rising in the other regions. SO_2/NO_2 in the economic zone of Western Sichuan Plateau showed a decreasing trend, while in the relatively clean ecological zone this ratio showed an increasing trend. Although the SO_2 pollution level in the clean areas was relatively low, the growth trend could not be ignored. The dependence of SO_2 and NO_2 on routine meteorological parameters in the ecological zone was low. The influence of meteorological parameters on gaseous pollutants was more significant in the economic zone with high anthropometric emissions.
- (2) O_3 pollution in the Western Sichuan Plateau with less artificial emission was prominent, with the highest annual mean concentration in Liangshan, $96.25 \pm 28.17 \mu\text{g}\cdot\text{m}^{-3}$, and the lowest annual mean concentration in Ganzi, $72.61 \pm 28.28 \mu\text{g}\cdot\text{m}^{-3}$, indicating that O_3 formation is promoted by strong solar radiation induced by smaller particle concentrations and more cloud-free days [42,43]. The pollution levels of SO_2 and NO_2 in the ecological zone were lower than those in the economic zone. The annual average concentration of SO_2 in Panzhihua and Aba was the highest and lowest, at $12.39 \pm 8.31 \mu\text{g}\cdot\text{m}^{-3}$ and $60.05 \pm 35.25 \mu\text{g}\cdot\text{m}^{-3}$, respectively. Panzhihua had the highest annual average NO_2 concentration, $37.81 \pm 11.98 \mu\text{g}\cdot\text{m}^{-3}$, while Aba had the lowest annual average concentration, $14.37 \pm 7.07 \mu\text{g}\cdot\text{m}^{-3}$. The seasonal and monthly variations of O_3 in the four regions were more obvious than those of SO_2 and NO_2 . The seasonal and monthly variations in the economic zone were higher than those in the ecological zone, indicating that man-made emissions have an important impact on the temporal distribution characteristics of air pollutants.

- (3) The correlation between SO₂ and NO₂ in Ganzi was the highest, followed by Panzhihua; the emission characteristics of SO₂ and NO₂ in these two regions were similar, and point sources contributed more to local air pollutants than mobile sources. The correlation between SO₂ and NO₂ was the lowest in Aba, indicating that the contribution of point sources to SO₂ and NO₂ pollution was not prominent. Ganzi showed the highest correlation coefficient, suggesting that NO₂ contribute the most to O₃ in this region in the Western Sichuan Plateau, followed by Panzhihua, Aba, and Liangshan. The correlation between NO₂ and O₃ is significantly higher in large and medium-sized cities in China than in the ecological and economic zones of Western Sichuan Plateau, which indicates that local precursors contribute more to the formation of O₃ in areas with high NO₂ emissions, while in areas with low NO₂ emissions, O₃ is more significantly affected by the regional background.

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