Original Research

Spatiotemporal Variations and Source of PM_{2.5} in the Sichuan Basin at Nanchong City, China

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> Received: 8 June 2024 Accepted: 9 September 2024

Abstract

To evaluate the pollution level, spatial and temporal variations and sources of fine particulate matter (PM, s) in Nanchong, a city in the Sichuan Basin, China, the trace element concentrations in PM, s were measured. The sources of PM, 5 were analyzed by the potential source contribution function (PSCF), enrichment factor (EF), and principal component analysis (PCA) methods from December 2014 to April 2016. The results showed that the mean concentrations of PM, s and the analyzed trace elements at the urban sites during the sampling periods were 58.89-63.03 µg m⁻³ and 2.81-3.43 µg m⁻³, respectively, and that those at the rural site were 60.13±4.28 µg m⁻³ and 2.55 µg m⁻³, respectively. There were no significant differences in the mean concentrations of PM25 or the analyzed trace elements between the urban area and the rural area. The seasonal variations in PM2.5 and the analyzed trace elements in the urban and rural areas were similar, with the order summer<spring<fall<winter. The PSCF analysis showed that the PM, s in the urban area of Nanchong City mainly originated from surrounding areas. The crustal elements K, Ca, Al, Na, Fe, Mg, and Ti were the dominant metals in the urban and rural areas of Nanchong, accounting for more than 93% of the total concentration of the analyzed trace elements. The EF of K was close to 20, indicating that it was derived mainly from anthropogenic sources. The EFs of Ca, Al, Na, Fe, Mg, and Ti were less than 10, indicating that these elements were derived mainly from natural sources. In contrast, although the concentrations of the trace elements Cu, Zn, As, Cd, Se, Ni, Pb, Ba, and Cr were relatively low, the EFs of each of these elements were much higher than 20, suggesting that these elements were mainly derived from anthropogenic sources. The EF and PCA results showed that the PM_{2.5} in Nanchong City mainly originates from natural sources, such as soil dust, and anthropogenic sources, such as biomass burning, construction dust, vehicular emissions, firework burning, etc.

Keywords: crustal elements, fine particulate matter, enrichment factor, principal component analysis, Nanchong City

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Introduction

In the past three decades, rapid economic development, urbanization, industrialization, population agglomeration caused by the siphoning effect of cities in China have caused serious atmospheric pollution [1-8]. When the new Ambient Air Quality Standard (GB 3095-2012) replaced GB 3095-1996 and was gradually implemented in China in 2013 [9], most regions and cities with excessively poor ambient air quality were affected by fine particulate matter (PM_{2.5}) particulate matter with an aerodynamic diameter less than 2.5 µm). Statistical assessments show that, in 2013, only 4.1% of the 74 important cities in China implemented new ambient air quality standards up to the PM_{2.5} standard [10]. After nearly ten years of continuous effort by governments at all levels, many experts and scholars, and people worldwide, PM25 pollution has been greatly alleviated, and only 25.4% of 339 cities in China had PM_{2.5} concentrations exceeding the national standard in 2022 [11]. Overtime, PM25 pollution, as the most important type of atmospheric pollution, has gradually become a public health concern and has become a research focus of scholars and experts because of its complex composition, wide range of pollution sources, influencing factors that vary from place to place, and great harm to human health [2, 4-5, 12-20].

Because the particle size of PM25 is small, it can float in the air for a long time. In contrast, PM_{2.5} has a large specific surface area and strong activity, and its surface can adsorb many carbonaceous components (sometimes called carbonaceous aerosols), watersoluble ions, and chemical elements [17, 21], which can then be carried into the human body through the respiratory tract and cause great harm to the human body, resulting in widespread scientific attention [19, 22]. PM_{2.5} mainly originates from natural sources (such as soil dust, [5, 23]) and anthropogenic sources (such as biomass burning, vehicle exhaust, firework burning, and construction dust, [5, 23-25]). In the atmosphere, once adsorbed to PM_{2.5}, elements are less likely to react with other substances, so they can remain adsorbed on the surface of PM_{2,5} for a long time. The contribution of each element to PM_{2.5} varies with different pollution sources, so the source of pollutants can be analyzed by identifying the characteristic elements in PM25. In the past, many experts and scholars have carried out many studies on the types, concentration levels, and sources of elements in PM_{2.5} and analyzed the sources of PM_{2.5} using models such as positive matrix factorization (PMF), chemical mass balance (CMB), and principal component analysis (PCA) [21, 25-26]. However, China has a vast territory, variable terrain, and different levels of economic development, and the developed regions (such as the Beijing-Tianjin-Hebei region [5, 19, 23, 27-29], the Yangtze River Delta region [5, 19, 23, 30-31], the Pearl River Delta region [5, 19, 23], etc.) and megacities [5, 32-40]) have attracted more attention than underdeveloped regions and smaller cities.

Nanchong City is a typical basin city, located in the northeastern Sichuan Basin and is the second most populous city in Sichuan Province. It is a typical agricultural city with fast development and a low level of industrialization. Its terrain slopes from north to south, with elevations between 888.8 and 256 meters. The urban area of the city is located in southern Nanchong City, with low elevations and extremely poor meteorological diffusion conditions for pollutants. The purpose of this study was to explore the pollution level, spatial and temporal variations, and sources of PM_{2.5} and elements in the urban and rural areas of Nanchong City to compensate for the shortage of domestic research on the sources and prevention of PM_{2.5} pollution in urban ambient air in the basin. The results of our study could provide insightful information and a scientific basis for the prevention and control of air pollution in basin cities and other types of Chinese cities in the future. The major objectives were as follows: (1) to determine the characteristics and sources of PM_{2.5}, (2) to analyze the seasonal variations in PM_{2.5} in urban areas and rural areas, (3) to determine the degree of element enrichment in PM_{2.5}, and (4) to determine the sources of elements in PM_{2,5}.

Materials and Methods

Study Area

Nanchong City (30°35′-31°51′N, 105°27′-106°58′E, 256-889 masl) is in the northeastern Sichuan Basin and in the midstream section of the Jialing River (Fig. 1a-b). This city has a mid-subtropical humid monsoon climate with four distinct seasons and is famous throughout the country for its poor meteorological diffusion conditions for pollutants, notably its breezy, damp, rainy, and foggy conditions. The landform type is mainly hilly, and the terrain slopes from north to south [41]. The urban area of Nanchong City is in the southern region and is at low elevations, i.e., approximately 300 meters (Fig. 1c). There are three districts in the main urban area of Nanchong, namely, Shunqing (SQ), Gaoping (GP), and Jialing (JL), with populations of more than 1.7 million and an area of approximately 170 km². In this study, one site was selected for PM_{2.5} sample collection in each of the three districts and in a rural area (the rural site is in Guihua Township (GH)) located approximately 25Kilometers from the urban area in the dominant upwind direction. The sampling site at SQ was situated on the rooftop (approximately 27 m above the ground) of the Nanchong Housing and Urban-Rural Development Bureau office building; at GP, it was situated on the rooftop (approximately 23 m above the ground) of the Gaoping Ecological and Environmental Bureau of Nanchong City office building; and at JL, it was situated on the rooftop (approximately 30 m above the ground) of the Jialing District People's Government's office building. The SQ, GP, and JL sampling sites were located

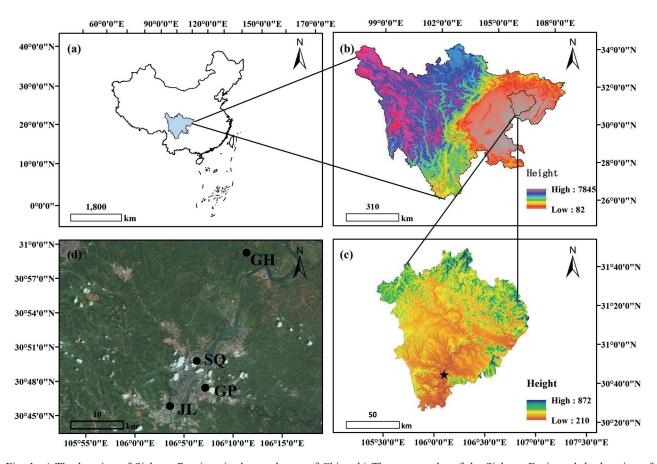


Fig. 1. a) The location of Sichuan Province in the southwest of China. b) The topography of the Sichuan Basin and the location of Nanchong in the northeastern Sichuan Basin. c) The topography of Nanchong. d) The Shunqing (SQ), Gaoping (GP), Jialing (JL), and Guihua (GH) sampling sites in Nanchong.

in mixed educational, commercial, traffic, and residential areas, and no obvious industrial pollution sources were detected within 3 km. Thus, the SQ, GP, and JL sampling sites can represent the urban environments of the Shunqing District, Gaoping District, and Jialing District, respectively. The sampling site at GH was located on the rooftop of the Guihua Township People's Government office building (approximately 8 m above the ground); this sampling site was surrounded by woodland, farmland, and villages without industrial pollution sources. The PM_{2.5} sampling sites are shown in Fig. 1d).

Sampling and Analysis

The PM_{2.5} samples were collected synchronously at the SQ, GP, JL, and GH sampling sites from December 22, 2014, to April 27, 2016. The PM_{2.5} samples were collected every six days with a four-channel sampler (each channel maintained a flow rate of 16.7 L min⁻¹) made by Tianhong Instrument Co., Ltd., of China (TH-16A), and the duration of each sampling was 24 hours (from 10:00 a.m. to 10:00 a.m. of the next day). A total of 86, 65, 74, and 89 samples were collected at SQ, GP, JL, and GH, respectively (Table S1). The PM_{2.5} samples were collected on Teflon and quartz

filters. The procedures for membrane replacement and sample storage were conducted following the Technical Guidelines for Source Analysis and Monitoring of Atmospheric Particulates (Trial) [42].

The Teflon filters were weighed at least three times with an electronic balance made by Mettler-Toledo Intl., Inc., USA (AX105 DeltaRange) after being balanced for 24 h in a super-clean environment at a constant temperature (20±1°C) and relative humidity (40%±3%). The PM_{2.5} concentration was obtained by dividing the mass difference of the Teflon filter before and after sampling by the sampled air volume.

The Teflon filters were nitrated in a solution of 3 mL HNO₃ (65%), 1 mL HCl (38%), and 0.2 mL HF (48%) at 175°C for 1 hour. The nitrification liquids were cooled to room temperature and diluted to 100mL with ultrapure water, and the elemental concentrations (Na, Mg, Al, Ca, Fe, K, Mn, Cu, Zn, As, Ti, Cd, Se, Ni, V, Pb, Co, Ba, and Cr) in the solutions were analyzed via inductively coupled plasma—mass spectrometry (Agilent 7500c, Agilent Technologies Co., Ltd., USA).

The hourly concentrations of PM_{2.5} and PM₁₀ were used to calculate and evaluate the proportion of PM_{2.5} in PM₁₀. The tracked aerological data for air masses (such as pressure and wind speed) for backward trajectory analysis were obtained from the Air Resource

Sampling sites	Spring	Summer	Autumn	Winter	Total
SQ	24	15	14	33	86
GP	19	15	13	18	65
JL	17	16	8	33	74
GH	20	15	13	41	89

Table S1. The numbers of samples of PM, s at SQ, GP, JL, and GH.

Laboratory, National Oceanic and Atmospheric Administration (http://www.arl.noaa.gov/). The hourly concentrations of PM_{2.5} and the tracked aerological data were used to calculate the potential source contribution function (PSCF).

Data Processing and Statistical Analysis

Potential Source Contribution Function (PSCF)

In this study, Metewoinfo software (download website: http://www.meteothinker.com) was used to analyze the 24-h backward trajectories of 500-m-high air masses in the urban area of Nanchong (30.80 N, 106.08 E, 500 m AGL) [43]. Then, the PSCF model in MetewoFo software, based on the results of backward trajectory analysis, was used to identify the potential source regions of PM, 5.

PSCF analysis relies on the conditional probability function as the basic principle and uses air mass trajectories to analyze and calculate the geographical location and spatial distribution of potential source areas. The research region is equally divided into $i \times j$ grid cells, and the ij^{th} cell is determined to affect the pollutant at the research site on the basis of the set pollutant concentration threshold. The PSCF_{ij} value in the ij^{th} cell is calculated by M_{ij}/N_{ij} , and grids with high PSCF $_{ij}$ values are considered potential source regions [35, 44]. The calculation formula of PSCF $_{ij}$ is as follows:

$$PSCF_{ij} = \frac{M_{ij}}{N_{ij}}$$
 (1)

where PSCF_{ij} denotes the ratios of M_{ij} and N_{ij} , M_{ij} denotes the number of trajectories that exceed the pollutant concentration threshold passing through the ij^{th} cell, and N_{ij} denotes the total number of trajectories of the air mass passing through the ij^{th} cell. In this study, the resolution of the grid cells was set to $1^{\circ} \times 1^{\circ}$, and the seasonal average concentration of $\operatorname{PM}_{2.5}$ was set as the threshold.

Uncertainties exist due to the small values of N_{ij} . Therefore, the weight function (W_{ij}) is used to reduce uncertainty [45]. The weight potential source contribution function (WPSCF) is then defined as the PSCF result multiplied by W_{ij} . The calculation formula of W_{ij} is as follows:

$$W_{ij} = \begin{cases} 1 & N_{ij} > 3N_{avg} \\ 0.72 & 1.5N_{avg} < N_{ij} \le 3N_{avg} \\ 0.4 & N_{avg} < N_{ij} \le 1.5N_{avg} \\ 0.2 & N_{ij} \le N_{avg} \end{cases}$$
(2)

where N_{avg} denotes the average number of endpoints of the air mass trajectories in the grid cells in the study area.

Enrichment Factor (EF)

The enrichment factor (EF) was used to analyze the relative contributions of anthropogenic sources and natural sources to the metal elements in PM_{2.5} [41, 46]. The calculation formula for the EF is as follows:

$$EF_{i} = \frac{(C_{i}/C_{ref})_{PM2.5}}{(C_{i}/C_{ref})_{soil}}$$
(3)

where EF_i denotes the enrichment factor of element i, $(C_i/C_{ref})_{PM2.5}$ denotes the ratio of the concentration of element i to the concentration of the reference element ref in PM_{2.5}, and $(C_i/C_{ref})_{soil}$ denotes the ratio of the concentration of element i to the concentration of the reference element ref in topsoil.

The evaluation criterion of the EFs of elements in $PM_{2.5}$ refers to [41, 47]. When the EFi of element i is greater than 10, an element is mainly contributed by an anthropogenic source. In contrast, when EFi is less than 10, the element is contributed mainly by a natural source. The concentrations of the elements in the topsoil were derived from the background values of the topsoil in Sichuan Province [48]. During the sampling period, the average concentration of iron (Fe) at the four sites was lower than that of aluminum (Al), and the concentration of Fe in the $PM_{2.5}$ in the GH was the lowest (Table S2). Therefore, Fe was selected as the reference element, and the Fe in $PM_{2.5}$ at the GH was used to calculate the EFs of the elements in $PM_{2.5}$ at the four sites.

Principal Component Analysis (PCA)

PCA is a multivariate analysis method that can be used as a source apportionment tool to explain the variance in the data effectively and identify the underlying reasons. In this study, the elemental data

Table S2. The background concentrations of topsoil elements in Sichuan Province. The data cited in Background values of soil elements in China (China National Environmental Monitoring Centre, 1990).

Elements	Mean	Unit
Na	0.85	%
Mg	0.85	%
Al	6.26	%
Са	1.13	%
Fe	3.3	%
K	2.02	%
Mn	657	mg/kg
Cu	31.1	mg/kg
Zn	86.5	mg/kg
As	10.4	mg/kg
Ti	0.4	%
Cd	0.079	mg/kg
Ti	0.4	%
Ni	32.6	mg/kg
V	96	mg/kg
Pb	30.9	mg/kg
Со	17.6	mg/kg
Ba	474	mg/kg
Cr	79	mg/kg

for PM_{2.5} at SQ, GP, JL, and GH during the sampling period were used to quantify the major sources of elements in PM_{2.5} and the contributions of each site via PCA. PCA was performed using SPSS 22.0 statistical software.

Data Processing and Analysis

The PM_{2.5} and element data were analyzed using the statistical software SPSS 22.0. The normality and homoscedasticity of the variables were checked via the Shapiro–Wilk and Levene tests, respectively. Normal and normalizable data were compared via one-way ANOVA (with Duncan's multiple range test). Nonparametric tests (with the Kruskal–Wallis H test) were used when the variables could not be normalized. The values are presented as the means±SEs. Spearman's correlation analysis was used to analyze the correlations between elements.

Origin-pro 9.1 and ArcGIS 10.8 software were used for data processing and graph construction.

Results and Discussion

Overview of PM_{2.5}

During the sampling period, the daily concentrations of PM_{2.5} at SQ ranged from 6.87 to 158.45 μg m⁻³, with 32.58% of daily PM_{2.5} concentrations exceeding the standard. The daily concentrations at GP ranged from 12.48 to 217.54 μg m⁻³, with 22.73% of daily PM_{2.5} concentrations exceeding the standard. The daily concentrations at JL ranged from 12.19 to 191.07 µg m⁻³, with 29.49% of daily PM_{2.5} concentrations exceeding the standard. Finally, the daily concentrations at GH ranged from 8.23 to 210.22 μg m⁻³, with 31.87% of daily PM_{2.5} concentrations exceeding the standard. The upper limit of the daily average PM_{2.5} concentration is 75 μg m⁻³ for Class II areas in the Standard of China's National Air Quality Standard, such as urban residential, commercial and traffic, cultural, industrial, and rural areas) [9]. The average concentrations of PM_{2.5} were 63.03±4.20 µg m⁻³ in SQ, 58.89±4.80 µg m⁻³ in GP, 61.70±4.41 µg m⁻³ in JL, and 60.13±4.28 µg m⁻³ in GH, which were slightly lower than those reported in Chengdu (67.0±43.4 μg m⁻³) and Chongqing (70.9±41.4 μg m⁻³) in the Sichuan Basin [49] but significantly lower than those reported in other cities in China (P<0.001), such as Beijing [28-29], Tianjin [28], Shijiazhuang [28], Chengde [28], Shanghai [30], Nanjing [30-31], Hangzhou [30], Ningbo [30], and Wuhan [34-35]. During the study period, the average concentrations at the four sites were significantly greater than that of the Grade II Standard of China's National Air Quality Standard for PM25 based on a one-sample t test (SQ, t = 6.682, P < 0.001; GP, t = 4.976, P < 0.001; JL, t = 6.051, P < 0.001; GH, t = 5.865, P < 0.001) (the upper limit of the annual average is 35 μg m⁻³ for Class II areas) [9], exceeding the standard by 0.80 times (SQ), 0.68 times (GP), 0.76 times (JL), and 0.72 times (GH). In addition, there was no significant difference in the concentration of PM_{2.5} among the four sites ($\chi^2 = 0.666$, P = 0.881), indicating that the PM_{2.5} pollution in the urban and rural areas of Nanchong City was relatively severe during the study period. Lei et al. [50] reported that PM_{2.5} was the major standard-exceeding pollutant in the urban area of Nanchong and that the mean annual concentrations of PM_{2.5} exceeded the Grade II Standard of China's National Air Quality Standard for PM, 5 (35 μg m⁻³) from 2015 to 2018. In other cities in the Sichuan Basin, the mean concentrations of PM_{2.5} ranged from 37 µg m⁻³ to 73 µg m⁻³, and the proportion of pollution caused by PM25 accounted for 77.83% of the total pollution from 2015 to 2016 [51]. These findings indicate that other cities in the Sichuan Basin also suffer from severe PM_{2.5} pollution.

Seasonal Variations in PM_{2.5}

The seasonal variations in the $PM_{2.5}$ concentration at the four sites are depicted in Fig. 2. The concentrations of $PM_{2.5}$ at the four sites were highest in winter, with mean

values of $94.06\pm7.07~\mu g~m^{-3}$ in SQ, $94.32\pm10.98~\mu g~m^{-3}$ in GP, $90.29\pm6.40~\mu g~m^{-3}$ in JL, and $87.25\pm6.05~\mu g~m^{-3}$ in GH, followed by spring and fall, with the lowest

concentrations in summer. Compared with those in other seasons, the sources of pollutants in Nanchong City did not change significantly in winter, but the mass

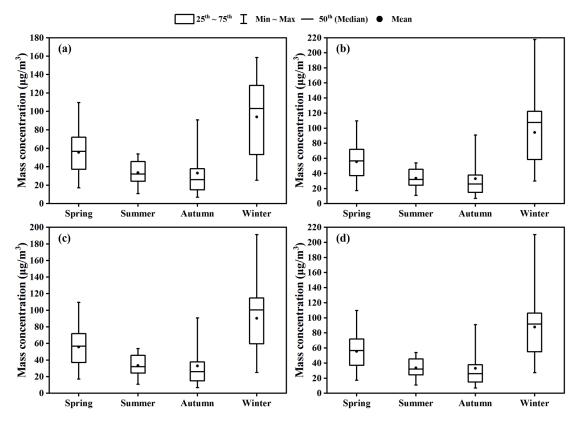


Fig. 2. The seasonal variations of PM_{2.5} concentration in SQ a), GP b), JL c), and GH d).

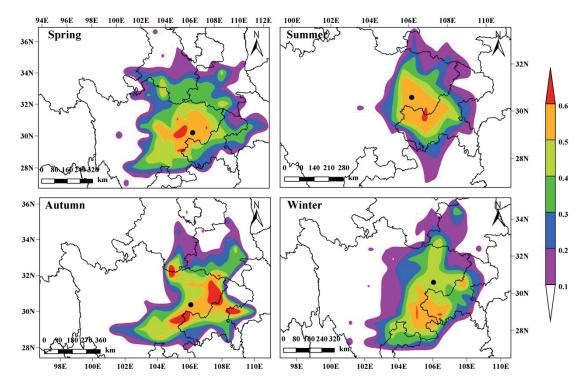


Fig. 3. Seasonal variation of WPSCF values of PM_{2.5} in the urban area of Nanchong City. ●, the location of the urban area of Nanchong City.

concentrations of PM_{2.5} increased significantly (SQ, $\chi^2 = 37.469$, P < 0.001; GP, $\chi^2 = 24.025$, P < 0.001; JL, $\chi^2 = 38.492$, P < 0.001; GH, $\chi^2 = 43.429$, P < 0.001). Similar seasonal trends have also been reported in other cities in the Sichuan Basin, such as Chengdu [38, 52-53], Chongqing [49], Leshan [38, 53], Dazhou [38, 53], and Ya'an [53-54]. Studies have shown that the concentration of particulate matter in the urban atmosphere is influenced by source emission intensities [20-21, 49, 55-56], atmospheric processes [49, meteorological conditions [49-50, 52, 58]. Yang et al. [58] reported that the low-level ventilation volume in cities in the Sichuan Basin is negatively correlated with the PM_{2.5} concentration and that a low wind speed can reduce the diffusion and migration of PM25, whereas temperature and relative humidity are favorable conditions for secondary aerosol generation.

As shown in Fig. 3, the mass concentration of PM_{2.5} in the urban area of Nanchong City was affected mainly by the surrounding areas because of poor meteorological diffusion conditions for pollutants (such as low wind speeds and high frequencies of calm winds) and advantageous conditions for the generation of PM_{2.5} (such as high temperatures and relative humidity). The proportion of the mass concentration of aerosols in Nanchong from the local and surrounding areas was 79.45% in spring, 85.42% in summer, 83.89% in fall, and 81.12% in winter [41]. In addition, Qiao et al. [59] reported that emissions within the Sichuan Basin contributed approximately 80% of the total PM_{2.5}

in Chengdu and Chongqing during the four seasons. Xu et al. [60] also confirmed that PM₂₅ in Chengdu in winter was less affected by the long-range transport of air masses, and the contribution of the long-traveled air masses was only 12.9%. Although this seasonal trend in PM_{2.5} concentrations has been widely found in cities in northern China [24, 35, 61], the backward trajectories of PM_{2,5} concentrations were completely different from those of cities in the Sichuan Basin, and the contribution of local pollution sources to PM_{2.5} from the end of December 2013 to the end of January 2014 was only 12.9% in Beijing and only 38.7% in Changchun [60]. Local pollution sources contributed no more than 60% of the total PM_{2.5} in Beijing's atmosphere during the whole heating season, and the contribution proportion of local pollution sources was only 40% in the summer [62].

Characteristics of the Elements

Nineteen elements in PM_{2.5} (Na, Mg, Al, Ca, Fe, K, Mn, Cu, Zn, As, Ti, Cd, Se, Ni, V, Pb, Co, Ba, and Cr) were measured in this study. During the sampling period, the total concentrations of the analyzed trace elements at SQ ranged from 0.45 to 20.45 μg m⁻³, with a mean value of 2.81±0.26 μg m⁻³; those at GP ranged from 0.53 to 49.00 μg m⁻³, with a mean value of 3.13±0.73 μg m⁻³; those at JL ranged from 0.41 to 37.36 μg m⁻³, with a mean value of 3.43±0.62 μg m⁻³; and those at GH ranged from 0.15 to 19.02 μg m⁻³, with s

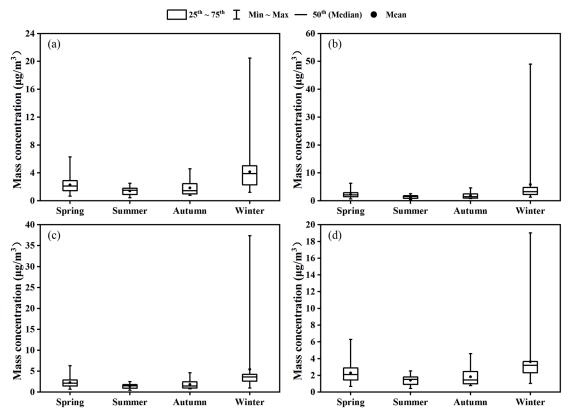


Fig. 4. The seasonal variations of the total concentration of the analyzed trace elements in SQ a), GP b), JL c), and GH d).

mean value of 2.55±0.26 μg m⁻³. These nineteen elements accounted for 1.43%-15.10% of the total mass of PM_{2.5} in SQ (mean value of 4.84%±0.25%), 1.73%-22.53% of the total mass of PM_{2.5} in GP (mean value of 4.83%±0.39%), 1.38%-26.33% of the total mass of PM_{2.5} in JL (mean value of 5.05%±0.42%), and 1.77%-15.73% of the total mass of PM_{2.5} in GH (mean value of 4.55%±0.25%). The total concentrations of the analyzed trace elements $(\chi^2 = 1.163, P = 0.762)$ and their mass fractions in PM_{2.5} $(\chi^2 = 0.816, P = 0.846)$ did not significantly differ among the four sites. Compared with those in other cities in China, the total concentrations of the analyzed trace elements in the PM_{2.5} in the urban area of Nanchong City were similar to those in Chengdu [37], Chongqing [37], and Jinan [39] but significantly lower than those in Beijing [28-29], Tianjin [28], Shijiazhuang [28], Chengde [28], Shanghai [30], Hangzhou [30], and Wuhan [34-35] (Table S3). Similarly, the total concentration of elements in PM₂₅ in the rural area of Nanchong was lower than that in the suburbs of Beijing [29], Ningbo [30], and Wuhan [29] (Table S3).

Furthermore, there was a significant positive correlation between the total elemental concentration in PM_{2.5} and the mass fraction of the 19 elements in PM_{2.5} among the four sites (P<0.001). However, the seasonal differences in the total concentrations of the analyzed trace elements (SQ, χ^2 = 31.447, P<0.001; GP, χ^2 = 22.702, P<0.001; JL, χ^2 = 26.281, P<0.001; GH, χ^2 = 38.962, P<0.001) and the mass fraction of the 19 elements in PM_{2.5} (SQ, χ^2 =10.099, P=0.018; GP, χ^2 =9.717, P<0.021; JL, χ^2 =7.976, P<0.047; GH, χ^2 =21.846, P<0.001) at

the four sites reached significant levels. Fig. 4 shows the seasonal variation trends of the analyzed trace elements at the four sites in Nanchong. The seasonal trends of the analyzed trace elements at the four sites were the same as those in PM_{2.5}, with the highest occurring in winter, intermediate values occurring in spring and autumn, and the lowest occurring in summer. However, the seasonal variation trends in the mass fractions of the analyzed trace elements in PM_{2.5} at the four sites were not the same. The mass fractions of the analyzed trace elements in PM_{2,5} at sites GP and GH were the highest in autumn, intermediate in spring and winter, and lowest in summer (Fig. 5b and 5d). In contrast to those in GP and GH, the mass fractions of the analyzed trace elements in PM25 ranked in the order autumn>summer>winter>spring at site SQ (Fig. 5a) and in the order autumn>winter> summer>spring at site JL (Fig. 5c).

Enrichment Factor (EF)

Fig. 6 shows the mean concentrations of the 19 elements in PM_{2.5} at the four sites during the sampling period. Generally, the distributions of the 19 elements in PM_{2.5} were similar among the four sites. K was found to be the dominant element in PM_{2.5} at the four sites, with K concentrations of 1.24±0.16 μg m⁻³ in SQ (Fig. 6a), 1.51±0.56 μg m⁻³ in GP (Fig. 6b), 1.62±0.44 μg m⁻³ in JL (Fig. 6c), and 1.10±0.15 μg m⁻³ in GH (Fig. 6d). The contribution of K to the total concentration of the analyzed trace elements in PM_{2.5} was 40.62%±1.21% in SQ, 38.06%±1.60% in GP, 38.40%±1.55% in

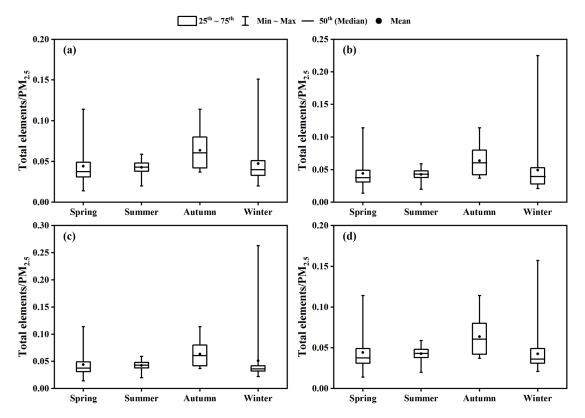


Fig. 5. The seasonal variations of the mass fraction of the analyzed trace elements in PM, 5 in SQ a), in GP b), in JL c), and GH d).

Table S3. The mean concentrations of elements in PM_{23} at SQ, GP, JL, GH, and the major cities of China. (unit: ng/m^3).

Reference	This study	This study	This study	This study	(Wang et al., 2018a)	(Wang et al., 2018a)	(Li et al., 2019)	(Li et al., 2019)	(Zhao et al., 2013)	(Ming et al., 2017)	(Ming et al., 2017)	(Yu et al., 2019)	(Ming et al., 2017)	(Ming et al., 2017)	(Xia et al., 2020)	(Acciai et al., 2017)	(Zhang et al., 2015)			
Co	0.16	0.18	0.17	0.12			0.59	0.31	0	0	0	0							3.51	
Сд	1.23	1.11	1.22	1.02			2.6	2.0	0	0	10	0			66.9			1.9		4.27
ï	1.25	4.11	2.41	1.08	2.1	4.2			10	10	10	0	14.9	9.3	3.62	10.4	8.25	6.0	3.39	4.8
As	1.84	2.20	1.94	2.80	10.8		10	9.5	30	20	20	10			11.9				27.91	34.71
>	2.06	2.44	1.77	2.95	1.9		2.6	1.2	0	0	0	10	16.5	9.88	6.05	15.3	7.36		9.01	
Se	3.99	3.57	3.54	2.97											5.15				8.1	6.55
Cr	7.00	16.68	11.17	12.64	5.6	11.1	9.5	2.0	20	10	10	10	16.9	13.2	00.9	17.9	14.6	5.2	13.93	7.91
Cu	12.59	9.31	14.08	10.28	18.7	11.3	49.8	23.1	40	140	40	20	24.2	24.7	27.2	38.5	16.2	16.8	23.26	25.27
Mn	16.02	14.38	15.83	10.24	33.8	37.7	59.3	37.4	70	100	120	09						29.1	50.94	97.25
Ti	18.28	18.29	18.43	13.99	32.5	46.5			40	40	40	80			25.9			26.3		
Ba	30.39	35.38	54.45	21.83					20	30	40	20			13.8			20.3	79.17	31.38
Pb	41.78	39.95	50.23	31.96	55.4	50.4	147.1	6.66	140	220	300	110	2.69	6.06	50.8	122	56.3	64.1	128.79	158.4
Zn	58.09	62.08	61.39	41.48	238	113	8.162	180.7	320	750	089	280	215	247	199	495	190	116.4	388.18 1	290.78
Mg	124.26 5	126.82 6	173.03 6	88.06 4			160.7	167.6	969	069	640	380	298	509	6.81	446	216	167.9	38	790 29
					9	9									4				.54	
Fe	3 227.44	8 248.84	5 234.36	1 146.70	456	989	1360.9	9.992	1490	2020	1840	1280	1340	942	577	1720	1160	655.1	1458.54	1680
Na	263.33	260.48	241.46	259.81														355		
Al	315.67	462.45	472.03	390.99	281	429	501	383.1	970	1170	1410	750	922	705		1370	1170	647.2		
Са	443.24	313.15	448.86	414.60	240	824	641.9	456.3	2420	3280	4270	1630	1930	1520	316	3020	1190	996.1	2043.36	4300
K	1237.51	1511.05	1623.54	1101.91	720	718	1789.9	718.6	1730	2150	3420	1150			610			648.7	3249.17	1530
Sampling site type	Urban	Urban	Urban	Rural	Urban	Urban	Urban	Suburban	Urban	Urban	Urban	Urban	Urban	Urban	Urban	Urban	Rural	Urban	Urban	Urban
Sampling sites	SQ	GP	JL	GH	Chengdu	Chongqing	Beijing	Beijing	Beijing	Tianjin	Shijiazhuang	Chengde	Shanghai	Nanjing	Nanjing	Hangzhou	Ningbo	Jinan	Wuhan	Wuhan

Wuhan	ourban	1740	5610		1970	910	350.61	910 350.61 203.00 32.98	32.98	82.86	82.86 31.45 11.74 7.46	74 7.46	49.37 6.40 5.51	10 5.51	(Zhang et al., 2015)	
Chengdu: The sam	pling site	e (Sichua	in Academy of	Environme	ntal Scienc	e) locate	d in the n	nixed area	of traffic, cc	mmerce, re	sidence, a	nd educati	on in Chengdu Ci	ity, Sichua	nce) located in the mixed area of traffic, commerce, residence, and education in Chengdu City, Sichuan Basin of southwest	
China. The sampli	ng campa	aign was	divided into for	ur periods:	autumn (23	3 October	r to 18 No	ovember,	2014), winte	r (6 January	to 2 Febr	1 2015 Jary, 2015), spring (2 to 29	April, 20	23 October to 18 November, 2014), winter (6 January to 2 February, 2015), spring (2 to 29 April, 2015), and summer (2 to	

30 July, 2015).

Chongqing: The sampling site (Chongqing Monitoring Center) located in the mixed area of traffic, commerce, and residence in Chengdu City, Sichuan Basin of southwest China. The sampling The sampling site (Institute of Atmospheric Physics, Chinese Academy of Sciences) located in the mixed area of traffic, residence, and education in Beijing City, China. The sampling campaign was divided into four periods: autumn (23 October to 18 November, 2014), winter (6 January to 2 February, 2015), spring (2 to 29 April, 2015), and summer (2 to 30 July, 2015).

The sampling site (the campus of the University of Chinese Academy of Sciences) located in the suburban area of Beijing City, China. The sampling period was from September 15 to period was from September 15 to November 12, 2014. November 12, 2014. Beijing: The sampling site (local meteorological bureau) located in the mixed area of traffic, commerce, and residence in Beijing City, north China. The sampling campaign was divided into four periods: spring (6 April to 1 May 2009), summer (9 July to 4 August 2009), autumn (11 October to 4 November 2009), and winter (14 January to 8 February 2010)

Shijiazhuang: The sampling site (local meteorological bureau) located in the mixed area of traffic, commerce, and residence in Shijiazhuang City, north China. The sampling campaign was divided l'anjin: The sampling site (local meteorological bureau) located in the mixed area of traffic, commerce, and residence in Tianjin City, north China. The sampling campaign was divided into four periods: spring (6 April to 1 May 2009), summer (9 July to 4 August 2009), autumn (11 October to 4 November 2009), and winter (14 January to 8 February 2010).

Chengde: The sampling site (local meteorological bureau) located in the mixed commercial and residential area in Chengde City, north China. The sampling campaign was divided into four into four periods: spring (6 April to 1 May 2009), summer (9 July to 4 August 2009), autumn (11 October to 4 November 2009), and winter (14 January to 8 February 2010) periods: spring (6 April to 1 May 2009), summer (9 July to 4 August 2009), autumn (11 October to 4 November 2009), and winter (14 January to 8 February 2010)

Shanghai: The sampling site located in the urban area of Shanghai City, east China. The sampling period was from September 2013 to August 2014.

Nanjing: The sampling site located in the urban area of Nanjing City, east China. The sampling period was from September 2013 to August 2014.

Nanjing: The sampling site (Nanjing Environmental Protection Building) located in the mixed area of traffic, commerce, and residence in Nanjing City, east China. The sampling period was from December 12, 2016 to December 31, 2017.

Hangzhou: The sampling site located in the urban area of Hangzhou City, east China. The sampling period was from September 2013 to August 2014.

Ningbo: The sampling site located in the rural area of Ningbo City, east China. The sampling period was from September 2013 to August 2014.

The sampling site (Jinan Environmental Monitoring Center Station) located in the mixed commercial and residential area in Jinan City, East China. The sampling period was from October 1, 2016 to September 30, 2017.

Wuhan: The sampling site (Super Monitoring Station of Wuhan) located in the typical residential urban area of Wuhan city, central China. The sampling period was from May 15 to May 31, 2014. Wuhan: The sampling site (the campus of the Wuhan University) located in the mixed area of traffic, commerce, residence and education in Wuhan city, central China. The sampling period was from August 2012 to July 2013.

Wuhan: The sampling site (Wuhan East Lake High-tech Zone) located in the suburban area of Wuhan city, central China. The sampling period was from August 2012 to July 2013 and August 2013 and A

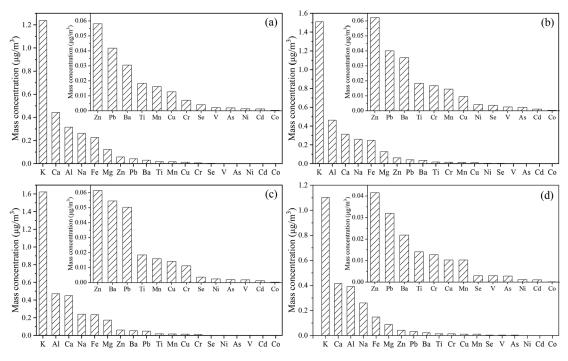


Fig. 6. The mass concentrations of the element in PM, 5 in SQ a), in GP b), in JL c), and in GH d).

JL, and 41.73%±1.651% in GH, and there was no significant difference among the four sites ($\chi^2 = 3.812$, P = 0.283). The crustal elements K, Ca, Al, Na, Fe, Mg, and Ti were the dominant metals at the four sites, and their contributions to the total concentration of the analyzed trace elements were 93.50%±0.20% in SQ, 93.01%±0.30% in GP, 93.03%±0.26% in JL, and 94.27%±0.21% in GH. Most studies have confirmed that crustal elements are the most important elements in PM_{2.5} and contribute 80.97%-92.73% of the total concentration of the analyzed trace elements in many Chinese cities (Table S3), such as Beijing [28-29], Chengdu [37], Chongqing [37], Tianjin [28], Hangzhou

[30], Jinan [39], Shijiazhuang [28], Ningbo [30], and Chengde [28].

EF analysis has been widely used to determine the source origins (anthropogenic or natural) of elements in PM_{2.5} [38, 47, 63]. K, Ca, Al, Na, Fe, Mg, and Ti were the most abundant elements in PM_{2.5} in Nanchong, and with the exception of the EF of K, which was close to 20, the EFs of the other crustal elements were very low. As shown in Fig. 7, the mean EF of K at the four sites was close to 20, indicating that K was derived mainly from anthropogenic sources. The mean EFs of the crustal elements Mg, Fe, and Al in PM_{2.5} at the four sites were significantly less than 5, indicating that they

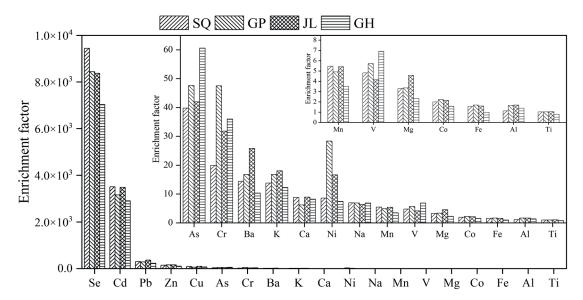


Fig. 7. The element enrichment factors in PM_{2.5} in SQ, GP, JL, and GH.

were derived mainly from natural dust. The mean EFs of the crustal elements Na and Ca and the trace element Ni were close to (or slightly greater than) 10, indicating that they were lightly enriched and influenced by both natural and anthropogenic sources. Unlike crustal elements such as K, Ca, Al, Na, Fe, and Mg, the crustal element Ti and the trace elements Co, V, and Mn have fewer anthropogenic sources, their concentrations were low, and their contributions to the total concentrations of the analyzed trace elements at the four sites were less than 1.10% during the sampling period. additionally, the EFs of Ti, Co, V, and Mn were less than 10, indicating that they were derived mainly from natural sources (Fig. 7). Nanchong is a typical agricultural city with rapid development and a low level of industrialization. Dust is emitted during the process of urban construction, resulting in a high concentration of crustal elements in PM, [50, 64-65]. In addition, owing to the lack of technology for processing straw and the high cost of processing straw, most straw has been burned directly in the past [50], which has contributed to the increase in the concentration of K in PM_{2.5}. Yang et al. [65] reported that the contribution rate of biomass burning to OC was greater than 60% in Nanchong City.

In contrast, the remaining 12 elements (Mn, Cu, Zn, As, Cd, Se, Ni, V, Pb, Co, Ba, and Cr) accounted for only 5.73%-6.99% of the total concentration of the analyzed trace elements in Nanchong, and the EFs of Co, V, and Mn were less than 10, suggesting that they were little influenced by anthropogenic sources. However, the EFs of Cu, Zn, As, Cd, Se, Ni, Pb, Ba, and Cr were much greater than 10, suggesting that these elements were mainly contributed by anthropogenic sources.

Source Apportionment via PCA

In this study, the sources of elements in $PM_{2.5}$ were estimated quantitatively via PCA with varimax rotation, and the retention of principal components with eigenvalues greater than 1.0 was used to identify the major pollutant sources. Fig. 8 shows the PCA results for the elements in $PM_{2.5}$ at the four sites.

At the SQ sampling site (the urban site), four principal component factors were found to explain the possible sources of elements in PM_{2.5} (Fig. 8a). Factor 1 (total variance 57.2%) was associated with elements such as Cd, Pb, Cu, K, Mn, and Zn. This group appears to represent a mixture of sources related to vehicular

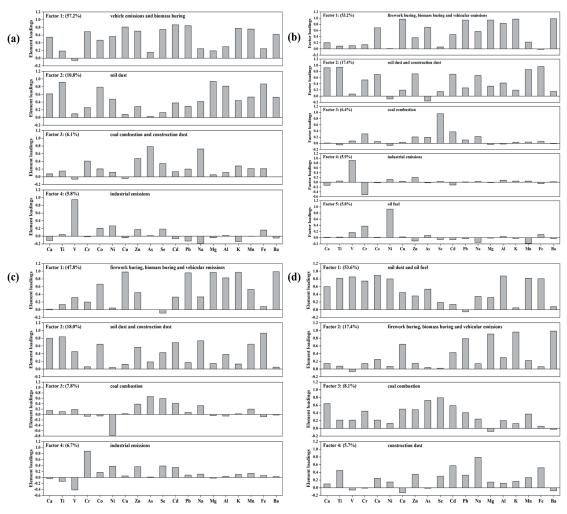


Fig. 8. Varimax-rotated principal component loading for elements in PM, s in SQ a), in GP b), in JL c), and GH d).

emissions [23, 66-67] and biomass burning [21, 25, 68-69]. Previous studies have shown that Cd is derived mostly from smelting and vehicular emissions [66, 70-71]; Pb and Cu are derived mostly from vehicular emissions [23, 66, 70] and industrial emissions [23, 72]; Zn is derived mostly from vehicular emissions, tire and brake debris, and industrial emissions [66, 73-74]; and Mn is derived from vehicular emissions, coal combustion, metal smelting [67] and soil dust [40, 73]. Cd ($EF_{Cd} = 3513.4$), Pb ($EF_{Pb} = 304.1$), Cu ($EF_{Cu} = 91.1$), Mn ($EF_{Mn} = 5.5$), and Zn ($EF_{Zn} = 151.1$) were significantly enriched and positively correlated with each other at the SQ sampling site (P < 0.01), indicating that they were derived mainly from vehicle emissions. Another element with a high degree of loading in factor 1, K, is derived mostly from biomass burning [21, 25, 68-69] and soil dust [40, 73]. The EF of K was 13.8 at the SQ sampling site during the sampling period, which was greater than 10 and much greater than that of other crustal elements (Ca, Al, Na, Fe, Mg, and Ti), indicating that K was strongly influenced by anthropogenic emissions. Therefore, the source of K in factor 1 was mainly biomass burning. The second factor (total variance: 10.8%) was assigned to emissions related to soil dust, a natural source. The crustal elements Mg, Ti, Fe, and Al, which had relatively high loadings in this factor, the EFs less than 5, indicating that they were derived from soil dust [40, 73]. The third factor included As and Na. This factor explains 6.1% of the total variance. It appears to be a mixture of sources related to coal combustion [27, 75-76] and construction dust [27, 77]. Se is frequently used as a marker of coal burning in many studies [75]. Although sodium hydroxide is often used in the exhaust gas desulfurization of coal-fired boilers, the final exhaust gas of coal-fired boilers often contains Na [77]. However, the concentration of Na was significantly greater than that of As at the SQ sampling site during the sampling period ($\chi^2 = 119.853$, P < 0.001), and the EF of Na was only 7.0, which was much lower than that of As $(EF_{4s} = 39.8)$, indicating that Na was less influenced by humans than As. In addition, the EF of Na was close to 10, indicating that Na was affected by certain human activities. Therefore, Na may be derived mainly from construction dust. Factor 4 explains 5.8% of the total variance in the data, with the highest loading of V and high loadings of Co, Se Zn, and Fe, representing contributions from industrial emissions [36].

At the GP sampling site (the urban site), five principal component factors were found to explain the possible sources of elements in PM_{2.5} (Fig. 8b). Factor 1 accounts for 53.2% of the total variance in the data, with Ba, K, Cu, Pb, Mg, and Al exhibiting the highest loadings, representing a mixture of sources related to firework burning [20, 65], biomass burning [21, 25, 68-69] and vehicular emissions [23, 66-67]. The raw materials of fireworks are black powders, and their major components are sulfur, charcoal powder, potassium chlorate, or potassium nitrate. Metal powders containing Cu, Mg, Al, and Ba are used as flame coloring and

glitter additives to add a festive effect. On February 9, 2016, the concentration of Ba was as high as 1.374 µg m⁻³, accounting for 0.63% of the total PM_{2.5}. In contrast, the concentration of Ba was only 0.015 in the non-Spring Festival period, accounting for only 0.03% of the total PM_{2.5}. While the concentrations and proportions of Cu, Mg, and Al were similar to those of Ba, their concentrations and proportions in PM, increased significantly during the Spring Festival compared with those during the non-Spring Festival. Factor 2 (17.4%). with high loadings of Fe, Ti, Ca, and Mn, represents soil dust [40, 73] and construction dust [27, 77]. Factor 3, with a high loading of Se, accounts for 6.4% of the total variance and represents coal combustion [27, 75-76]. Factor 4 accounts for 5.9% of the total variance in the data, with V exhibiting the highest loading and Zn and Ni exhibiting relatively high loadings, representing contributions from industrial emissions [36]. Factor 5, explaining 5.8% of the variance and exhibiting the highest loading for Ni and high loadings of Cr and V, is considered related to oil fuel [32-33].

At the JL sampling site (the urban site), four principal component factors were found to explain the possible sources of elements in PM_{2.5} (Fig. 8c). The metal sources at JL are similar to those at GP, and factor 1 accounts for 47.8% of the total variance in the data, with the highest loadings for Ba, Cu, K, Mg, Pb, and Al, representing a mixture of sources related to firework burning [20, 65], biomass burning [21, 25, 68-69] and vehicular emissions [23, 66-67]. Factor 2 (18.0%) is related to soil dust [40, 73] and construction dust [27, 77]. Factor 3 (7.8%) is related to coal combustion [27, 75-76] and is characterized by high loadings of As and Se. Factor 4 (6.7%), with the highest loading of Cr and high loadings of Se, Ni, Zn, and Cd, represents industrial emissions [36].

At the GH sampling site (rural site), four principal component factors were found to explain the possible sources of elements in PM_{2.5} (Fig. 8d). Factor 1 (total variance 53.6%) was assigned to emissions related to soil dust [40, 73] and oil fuel [32-33]. Al, Ti, Mn, and Fe had high loadings in this factor and are indicative of primary crustal sources. V and Ni are considered markers of oil fuel. Factor 2 accounts for 17.4% of the total variance in the data and has high loadings of Ba, K, Mg, and Pb, suggesting that firework burning [20, 65], biomass burning [27, 75-76], and vehicular emissions [27, 75-76] were the major contributors. Factor 3 accounts for 8.1% of the total variance in the data, with high loadings of As and Se representing contributions from coal combustion [27, 75-76]. A high loading of Na is observed in factor 4 (5.7%), which is considered related to construction dust

In general, the metal sources of PM_{2.5} at the urban sampling sites in Nanchong were consistent. Biomass burning, vehicular emissions, firework burning, soil dust, and construction dust were the most important sources of metals in PM_{2.5}, and the total contribution of these sources was greater than 65%. Additionally,

the contributions of coal combustion and industrial emissions were relatively low, while contributions from oil fuel were observed only at the GP sampling site. For the rural GH sampling site, the metal sources with the highest contributions to PM_{2.5} were soil dust, oil fuel, firework burning, biomass burning, and vehicular emissions, whereas the contributions of coal combustion and industrial emissions were lower. Nanchong is an agricultural city with a low level of industrial development. Owing to the lack of relevant straw treatment technology and the high cost of straw treatment, many stalks are directly burned in the open air after crop harvest, resulting in a sharp increase in the PM_{2.5} concentration. In addition, Nanchong City is developing rapidly, and it features a large population and a high number of motor vehicles. By the end of 2022, the number of motor vehicles in Nanchong was close to 1.2 million, and the total acreage and population of the main urban area were 171 square kilometers and 1.61 million, respectively. Therefore, vehicular emissions and construction dust are important sources of urban air pollution. In contrast, owing to the lower level of industrialization, the demand for coal in Nanchong has not been high, and since 2000, the main urban area of Nanchong has implemented the policy of replacing coal with gas. Therefore, the contributions of coal combustion and industrial emissions to PM_{2.5} were small.

Conclusions

During the study period, the average concentrations of PM_{2.5} in SQ, GP, JL, and GH were 63.03±4.20 µg m⁻³, 58.89±4.80 µg m⁻³, 61.70±4.41 µg m⁻³, and 60.13±4.28 µg m⁻³, respectively, which exceeded the Grade II Standard of China's National Air Quality Standard for PM_{2.5} by 0.80, 0.68, 0.76, and 0.72 times, respectively. The new Ambient Air Quality Standard (GB 3095-2012) replaced GB 3095-1996 and was implemented in China in 2014, and PM_{2.5} was a newly added monitoring item. During this period, China's economy was in a period of rapid development, and the control of PM_{2.5} in most cities was still in the exploratory stage. As a result, Nanchong, like other cities in China, faces serious PM_{2.5} pollution.

Nanchong City is located in the northeastern Sichuan Basin, it is low in elevation, and the meteorological conditions do not favor pollutant diffusion. Because of these poor meteorological diffusion conditions for pollutant dispersal (such as low wind speeds and high frequencies of calm winds) and advantageous conditions for generating PM_{2.5} (such as higher temperatures and relative humidity), the PM_{2.5} in Nanchong City mainly originates from surrounding areas. There was little difference in PM_{2.5} concentration between the main urban area and the rural areas, and the seasonal trends were the same, with the highest concentration occurring in winter, intermediate values occurring in spring

and autumn, and the lowest concentration occurring

The total concentrations of the analyzed trace elements in SQ, GP, JL, and GH were 2.81±0.26 μg m⁻³, $3.13\pm0.73 \,\mu g \, m^{-3}$, $3.43\pm0.62 \,\mu g \, m^{-3}$, and $2.55\pm0.26 \,\mu g \, m^{-3}$, respectively, and the mass fractions of those elements in $PM_{2.5}$ were 4.84%±0.25%, 4.83%±0.39%, 5.05%±0.42%, and 4.55%±0.25%, respectively. The total concentrations of the analyzed trace elements and their mass fractions in PM, did not significantly differ among the four sites. The seasonal variation trends of the analyzed trace elements at the four sites were consistent, with the highest occurring in winter, intermediate values occurring in spring or autumn, and the lowest occurring in summer. However, the seasonal variation trends of the mass fractions of the 19 elements in PM_{2.5} at the four sites exhibited the highest values in autumn and lowest in spring or summer.

The crustal elements K, Ca, Al, Na, Fe, Mg, and Ti were the most abundant in PM_{2.5} in Nanchong, and their contributions to the total mass of the analyzed trace elements were 93.50%±0.20%, 93.01%±0.30%, $93.03\% \pm 0.26\%$, and $94.27\% \pm 0.21\%$ at the SQ, GP, JL, and GH sites, respectively. The EF of K in Nanchong City was close to 20, indicating that it was mainly contributed by anthropogenic sources, and the EFs of other crustal elements were less than 10, indicating that they were derived primarily from natural sources. In contrast, the remaining 12 elements (Mn, Cu, Zn, As, Cd, Se, Ni, V, Pb, Co, Ba, and Cr) accounted for only 5.73%-6.99% of the total mass of the analyzed trace elements in Nanchong. Nevertheless, the EFs of these elements were each much greater than 50, suggesting that they were contributed mainly by anthropogenic sources.

Nanchong City has a large population, its economy is mainly agricultural, and its industrial development is relatively weak. The PCA results revealed that the PM_{2.5} in the urban area of Nanchong City mainly originates from natural sources such as soil dust and anthropogenic sources such as biomass burning, construction dust, vehicular emissions, and firework burning. In rural areas of Nanchong City, the PM_{2.5} mainly originates from soil dust, oil fuel, biomass burning, firework burning, and vehicular emissions. The contributions of coal combustion and industrial emissions to PM_{2.5} were lower in the urban and rural areas of Nanchong City.

Acknowledgments

The authors sincerely thank Nanchong Ecological and Environmental Monitoring Central Station of Sichuan Province for the permission to access their data. This study was supported by the Science and Technology Plan Project of Nanchong City (22YYJCYJ0001 and 23YYJCYJ0053). The opinions in this study do not reflect the views or policies of Nanchong Ecological and Environmental Monitoring Central Station of Sichuan Province.

Conflict of Interest

The authors declare that they have no conflict of interest.

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