Potential Risks of PM$_{2.5}$ in Urban and Suburban Environments: A Dual Perspective on Chemical Constituents and Pollution Sources

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ABSTRACT

To investigate the chemical characteristics and source profiles of fine particulate matter (PM$_{2.5}$) between suburban and urban areas, intensive observation and sample collection were conducted from November 2019 to January 2020 covering both clean and polluted days in Yulin, southwestern China. Higher pollution levels were observed in urban areas during both clean and polluted days. The positive matrix factorization (PMF) receptor model and the analysis of absolute principal component scores-multiple linear regression (APCS-MLR) revealed that mixed sources and vehicle exhaust were significant contributors in both suburban and urban areas. Vehicle exhaust (36.0%), combustion sources (22.0%), and crustal dust (20.3%) were found to be the main factors affecting air quality on polluted days. The backward trajectory and the potential source contribution function (PSCF) analysis indicated that the pollution primarily originated from local sources as well as neighboring cities. According to the geo-accumulation index ($I_{geo}$), As, Cu, Pb, Se, and Zn were identified as harmful species. Moreover, non-carcinogenic risks cannot be ignored, particularly for As and V. Inhalation exposure to combustion source is identified as the primary contributor to human health risks during both clean and polluted periods. Mixed sources and industry source were the main factors causing the carcinogenic risk between suburban and urban areas.

Keywords: Particle pollution, Chemical characteristics, Source profile, Regional transport, Health risks

1 INTRODUCTION

Environmental particulate matter is definitely dominant in air pollution, especially fine particles (particles with aerodynamic diameter $\leq 2.5$ μm, PM$_{2.5}$) (Lei et al., 2021; Rahman et al., 2020). The highly active PM$_{2.5}$ are often accompanied by toxic and hazardous substances, including heavy metals and microorganisms. These pollutants possess the capability to deeply infiltrate the alveoli and bronchiole, leading to respiratory and cardiovascular diseases that pose a significant threat to human health (Juda-Rezler et al., 2021; Rönkkö et al., 2020; Avise et al., 2009; Tai et al., 2010). China has implemented stricter air pollution control policies and has achieved remarkable progress in pollution control during the 13th Five-Year Plan (Wang et al., 2020; Xiao et al., 2020). In 2020, approximately 37.1% of cities in China still exceeded the prescribed PM$_{2.5}$ standard (with an average annual value was 35 μg m$^{-3}$), of which 24 cities exceeded the standard by more than 50%, and about 77% of heavy pollution was caused by PM$_{2.5}$ (Ministry of Ecology and Environment of the PRC). Currently, more research is dedicated to elucidating the mechanisms underlying the formation of severe haze pollution (Chang et al., 2021; Chen et al., 2020b; Dimitriou et al., 2021; Jeong et al., 2020; Li et al., 2020, 2021b). To further improve air quality and clarify its impact on human health, it remains imperative to persistently pay attention to the characteristics of PM$_{2.5}$
pollution and potential health risks across diverse cities at different time intervals (Shen et al., 2020; Yang et al., 2021).

The identification and quantification of emission source are crucial focal points in mitigating PM$_{2.5}$ concentrations. Aerosol-bound PM$_{2.5}$ is an intricate mixture derived from various natural and anthropogenic sources, including forest fires, soil and rock weathering, industrial emissions, vehicle exhaust and agricultural activities (Jia et al., 2022; Li et al., 2023; Luo et al., 2022). The chemical composition of PM$_{2.5}$ can serve as a tracer for pollution characteristics and source identification. High levels of carbonaceous species are observed in sources associated with coal combustion and vehicle emissions (Wu et al., 2020b; Yao et al., 2020). Regarding water-soluble inorganic ions, Ca$^{2+}$ is the typical tracer of fugitive dust (Li et al., 2021a). K$^{+}$ is usually employed as a marker for biomass burning (Dutta and Chatterjee, 2021; Uranishi et al., 2019). Trace metals (TMs) pose significant environmental contamination and potential health risks due to their chemical toxicity and tendency to accumulate (Huang et al., 2021; Jia et al., 2022; Liu et al., 2023). The exposure assessment based on receptor models can quantitatively identify sources that cause potential health impacts (Shen et al., 2021; Chen et al., 2017; Ji et al., 2019; Luo et al., 2021; Wang et al., 2021b). Limited research has been conducted on PM$_{2.5}$ variation in less developed areas. Especially in recent years, there have been periods of polluted weather that raised concerns. In addition to differences in industrial distribution, the influence of geographical characteristics and transmission routes should also be marked (He et al., 2017; Lei et al., 2021; Liu et al., 2021).

This study aims to provide a comprehensive understanding by investigating the variability and behavior of chemical components in PM$_{2.5}$ under diverse conditions, including urban and suburban areas, as well as clean and pollution periods. The application of PMF, APCS-MLR, and PSCF models facilitates source identification, apportionment, and assessment of their impact on human health risks. The results will help to improve the response mechanism for polluted weather.

2 MATERIALS AND METHODS

2.1 Study Area and Sample Collection

The sampling campaign was conducted from November 24th, 2019 to January 10th, 2020 in Yulin (109°39′–110°18′E, 22°19′–23°01′N), with a population of approximately 5.80 million residents and over 744 thousand civil vehicles as of 2020. All six observation sites were Environmental Monitoring Stations for routine air quality measurements, covering both suburban areas (SYDQ, FM, and HSSK) and urban areas (SJCZ, NJYZ, and CXEZ) (Fig. 1). The sampling frequency was set at three times per week, with daily sampling conducted during the pollution period. Each sample was collected for 24-hour. A total of 125 PM$_{2.5}$ samples were collected using median-volume air samplers (100 L min$^{-1}$, TH-150C III, Wuhan, China). Additionally, 1–2 blank samples were collected at each site during the observation period. The detailed treatment method of fiber filters is provided in Text S1.

2.2 Chemical Analysis and Observation Data

Elemental compositions (including Al, As, Ba, Ca, Co, Cr, Cu, Fe, K, Mg, Mn, Na, Ni, Pb, Se, Ti, V, and Zn) were determined using an inductively coupled plasma mass spectrometer (ICP-MS, PerkinElmer, NexION-350X, USA). Water-soluble inorganic ions (WSIs, including K$^+$, NH$_4^+$, SO$_4^{2-}$, and NO$_3^-$) were analyzed using an ion chromatograph (IC, DIONEX, Model ICS-1600, USA). The analysis of organic carbon (OC) and elemental carbon (EC) was conducted using the Desert Research Institute (DRI) Model 2015 Carbon analyzer (Atmoslytic Inc., Calabasas, CA), based on the Interagency Monitoring of Protected Visual Environment (IMPROVE-A) thermal/optical reflectance (TOR) protocol. The detailed methodology is outlined in Text S2.

In this study, air quality levels were obtained from the China National Environment Monitoring Center. Hourly data of atmospheric pollutants (PM$_{2.5}$, PM$_{10}$, SO$_2$, NO$_2$) and meteorological parameters, including temperature (T, °C), relative humidity (RH, %), wind speed (WS, m s$^{-1}$), and wind direction (WD, °), were obtained from the Yulin Air Quality Network Monitoring and Management Platform. Air mass information was procured from the Global Data Assimilation System (GDAS) within the National Center for Environmental Prediction (NCEP).
2.3 Index of Geo-accumulation

The assessment of heavy metal pollution levels utilized the geo-accumulation index (I_{geo}). I_{geo} can be calculated as follows (Kara et al., 2014; Kicińska and Wikar, 2021):

\[ I_{geo} = \log_2 \left( \frac{C_i}{B_i} \right) \]

where \( C_i \) represents the concentration of the \( i \)-th metal species in PM_{2.5} sample, \( B_i \) is the background concentration of the \( i \)-th metal species, and 1.5 serves as a correction factor to account for anthropogenic impact. The background values for each species are as follows: As (15 mg kg\(^{-1}\)), Ba (289 mg kg\(^{-1}\)), Co (8 mg kg\(^{-1}\)), Cr (73 mg kg\(^{-1}\)), Cu (24 mg kg\(^{-1}\)), Mn (181 mg kg\(^{-1}\)), Ni (23 mg kg\(^{-1}\)), Pb (24 mg kg\(^{-1}\)), Se (0.5 mg kg\(^{-1}\)), Ti (5256 mg kg\(^{-1}\)), V (114 mg kg\(^{-1}\)), and Zn (67 mg kg\(^{-1}\)).

2.4 Source Apportionment

In this study, two resolution methods were adopted for source apportionment. The positive matrix factorization (PMF) model, endorsed by the United States Environmental Protection Agency (U.S. EPA), was utilized to characterize PM_{2.5} sources, with a detailed methodological explanation provided in Text S3 (Chen et al., 2007; Zou et al., 2017).

Absolute principal component scores-multiple linear regression (APCS-MLR) receptor model was derived by Thurston and Spengler (1985). Considering different scenarios of spatiotemporal scale in this study, the APCS-MLR method was used to analyze the source apportionment during both clean period and pollution period. The first is to transform the observation data into standard form using the following equation (Wang et al., 2021a):

\[ Z_{ik} = \frac{C_{ik} - \bar{C}_i}{\sigma_i} \]

where \( Z_{ik} \) (Z-score) represents the standardized value of \( i \)-th component in the \( k \)-th sample; \( C_{ik} \) is the observed concentration of \( i \)-th component in the \( k \)-th sample; \( \bar{C}_i \) and \( \sigma_i \) are the mean value and standard deviation of the \( i \)-th component across all samples.

Secondly, the major factors are determined using the standardized value through the following equation:
\[ A_k = \sum_{i=1}^{n} w_{ij} \cdot z_{ik} \]  

where \( A_k \) represents the factor score of the \( j \)-th source to the \( k \)-th sample; \( w_{ij} \) is the coefficient matrix of the \( i \)-th component to the \( j \)-th source. Detailed methods for the implementation of APCS-MLR can be found in Text S4.

### 2.5 Potential Source Function (PSCF) Model

The PSCF values for the grid cells within the study domain are determined by tallying the endpoints of trajectory segments that terminate within each individual cell (Li et al., 2023; Zhan et al., 2021; Zong et al., 2018). The PSCF value for the \( ij \)-th cell is then defined as:

\[ PSCF_{ij} = \frac{m_{ij}}{n_{ij}} \]

where \( n_{ij} \) represents the total points in the \( ij \)-th cell; \( m_{ij} \) is the total points in the 75\(^{th} \) percentile threshold criterion in this study. The calculation of the PSCF value was adjusted by incorporating an arbitrary weight function \( W(n_{ij}) \) to mitigate the uncertainties (Polissar et al., 1999). The total number of endpoints in a specific cell was below three times the average value of endpoints per cell (Zong et al., 2018). \( W_{ij} \) was defined as follows:

\[
W_{ij} = \begin{cases} 
1.00 & (80 < n_{ij}) \\
0.70 & (20 < n_{ij} \leq 80) \\
0.42 & (10 < n_{ij} \leq 20) \\
0.05 & (n_{ij} \leq 100) 
\end{cases}
\]

### 2.6 Human Health Risk Assessment

According to the recommended method outlined by U.S. EPA (2009, 2014, 2018), inhalation exposure concentrations (EC) for each pollutant can be determined using the following equation:

\[ EC_{ij} = \frac{C_{ij} \times ET \times EF \times ED}{AT} \]

where \( EC_{ij} \) is the exposure concentration of \( i \)-th component in \( j \)-th source; \( C_{ij} \) is the estimated concentration of \( i \)-th component in \( j \)-th source; \( ET \) is the exposure time; \( EF \) is the exposure frequency; \( ED \) is the exposure duration; \( AT \) is the average time (Table S4).

Carcinogenic risk (CR) for each element (\( CR_i \)) can be calculated by Eq. (7), and the source contribution of carcinogenic risk for each source (\( SC(ca)_i \)) can be calculated by Eq. (8):

\[ CR_i = \sum_{j=1}^{n} EC_{ij} \cdot IUR_i \]  

\[ SC(ca)_i = \frac{CR_j}{\sum_{j=1}^{n} CR_j} = \frac{CR_j}{\sum_{j=1}^{n} \sum_{i=1}^{n} EC_{ij} \cdot IUR_i} \]

where \( IUR_i \) represents the inhalation unit risk associated with the \( i \)-th component (Table S5); \( CR_j \) is the carcinogenic risk attributed to the \( j \)-th source.
Hazard quotient (HQ) for each element ($HQ_i$) can be calculated by Eq. (9), and the source contribution of non-carcinogenic risk for each source ($SC(nca)_i$) can be calculated by Eq. (10):

$$HQ_i = \sum_{j=1}^{p} \frac{EC_{ij}}{RfC_i}$$  

$$SC(nca)_i = \frac{HQ_j}{\sum_{j=1}^{p} HQ_j} = \frac{HQ_j}{\sum_{j=1}^{p} \sum_{i=1}^{p} \frac{EC_{ij}}{RfC_i}}$$

where $RfC_i$ represents the reference concentration associated with $i$-th component (Table S5); $HQ_i$ is the non-carcinogenic risk for $j$-th source.

3 RESULTS AND DISCUSSION

3.1 Overview of Field Observations

3.1.1 Characteristics of air pollutants and meteorological parameters

Fig. 2 illustrates the characteristics of observed meteorological parameters and pollutants at representative sites, namely the SJCZ site in urban areas and the SYDQ site in suburban areas. The average concentrations of hourly PM$_{10}$, PM$_{2.5}$, SO$_2$, and NO$_2$ at the SJCZ site were $80.1 \pm 36.8$ µg m$^{-3}$, $49.1 \pm 21.9$ µg m$^{-3}$, $24.7 \pm 22.0$ µg m$^{-3}$, and $28.4 \pm 22.1$ µg m$^{-3}$, respectively; while at the SYDQ site they were recorded as $69.6 \pm 28.7$ µg m$^{-3}$, $44.0 \pm 21.7$ µg m$^{-3}$, $16.6 \pm 14.5$ µg m$^{-3}$, and $21.8 \pm 21.3$ µg m$^{-3}$, respectively.

Fig. 2. Time series of the hourly meteorological parameters, PM$_{10}$, PM$_{2.5}$, SO$_2$, and NO$_2$ during the observation period at representative sites in (a) urban and (b) suburban areas, respectively.
the concentrations of TMs increased by 2.7 μg m⁻³. At the SJCZ site, the values of WS and RH were 1.2 ± 0.7 m s⁻¹ and 51.6% ± 16.0%, respectively, while at the SYDQ site, they were 1.2 ± 1.1 m s⁻¹ and 62.5% ± 18.2%. According to the air quality index (AQI) categories, the entire observation period was divided into clean days (AQI ≤ 100) and PM₂.₅-polluted days (AQI > 100 with PM₂.₅ as the primary pollutant). During the pollution episode, the ranges of meteorological parameters showed clear differences. Lower WS was observed in polluted days (ranging from 0.1 m s⁻¹ to 2.4 m s⁻¹). During this period, the RH has a rising process.

Higher concentrations of pollutants were found on polluted days during the observations (Fig. S1). The concentrations of PM₁₀, PM₂.₅, SO₂, and NO₂ generally presented a similar variation tendency during pollution episode both at suburban and urban sites. During the pollution period, the NJYZ site exhibited the highest concentration of PM₁₀ (168.9 μg m⁻³), followed by the CXEZ site (161.6 μg m⁻³) and the SJCZ site (123.7 μg m⁻³), with an increase rate of 117.3%, 87.8%, and 73.6%, respectively, compared to the clean period. PM₂.₅ concentrations showed similar variability as PM₁₀, with increases ranging from 72.5% to 112.2% in urban sites and 43.8% to 71.0% in suburban sites during the pollution period. In addition, NO₂ concentrations increased the most during the pollution period, with increases ranging from 107.8% to 137.2% in urban sites and 47.7% to 94.3% in suburban sites. In comparison, urban areas exhibited higher levels of pollution than suburban areas in both clean and polluted days, with a greater increase rate of pollutants observed in urban areas during the pollution period. Additionally, the wind speed was lower during the pollution period, which hindered air diffusion and exacerbated PM₂.₅ pollution.

3.1.2 Characteristics of chemical composition in PM₂.₅

In Fig. 3, variations, and fractions of the main components in PM₂.₅ during both clean and polluted period is presented for suburban and urban sites, respectively. In comparison to the clean period, the concentration of OC, EC, TMs, and SNA (sum of SO₄²⁻, NO₃⁻, and NH₄⁺) during the pollution period increased by 7.6 μg m⁻³, 2.2 μg m⁻³, 2.8 μg m⁻³, and 22.7 μg m⁻³, respectively (Fig. 3(a)). Carbon species, including OC and EC, are pivotal components of PM₂.₅, often associated with various emission sources such as coal combustion, biomass burning, and traffic emissions (Chen et al., 2020a; Cheng et al., 2010; Liu et al., 2020). The OC/EC ratio, which is a tracer for identifying pollution sources, typically ranges from 0.3 to 7.6 for coal combustion and from 0.7 to 2.4 for vehicle emissions (Zeng et al., 2021). In this study, the OC/EC ratio ranged from 1.5 to 7.5 with an average value of 3.5 ± 0.9. During the clean period and pollution episodes, the OC/EC ratio was 2.8 (R² = 0.6) and 2.3 (R² = 0.7), respectively, suggesting that they closely originated from combustion sources and vehicle emissions (Fig. S2) (Li et al., 2023; Tao et al., 2018). The dominant species in PM₂.₅ were SNA, which exhibited a significant increase of 15.3% during the pollution period (Fig. 3(c)). Moreover, SNA concentrations increased by 14.9% at suburban sites and 15.5% at urban sites (Fig. 3(d)). The concentration of SO₄²⁻ showed the highest level in SNA and exhibited the largest increase during the pollution episode. Moreover, the fractions were similar compared to the components detected in the suburban and urban samples (Fig. 3(c)). The detected components generally exhibited higher concentrations in urban sites compared to suburban sites. Specifically, the concentrations of TMs increased by 2.7 μg m⁻³ at suburban sites and 3.0 μg m⁻³ at urban sites (Fig. 3(b)), while the ratio decreased 5.9% at suburban sites and 7.6% at urban sites during the pollution period (Fig. 3(d)).

The variations in concentration of the major components of PM₂.₅ at suburban and urban sites during periods of clean and pollution are presented for comparison in Table S1. Although elements contribute to a minor proportion of PM₂.₅ (approximately 10.2% in this study), they serve as crucial indicators for tracing pollution sources (Zong et al., 2018). The major elemental composition was Na, K, Fe, Ca, Al, Mg, and V. Additionally, higher concentration was observed for Zn (64.1 ± 34.4 ng m⁻³) during the sampling period, followed by As (58.3 ± 62.9 ng m⁻³), Pb (42.2 ± 36.4 ng m⁻³), Mn (29.6 ± 15.8 ng m⁻³), Cr (15.3 ± 9.9 ng m⁻³), and Cu (14.3 ± 9.0 ng m⁻³). The concentrations of As and Zn notably increased during the pollution period, indicating significant contributions from anthropogenic activities involving industrial emissions, vehicle exhaust and combustion emissions (Jing et al., 2016; Lei et al., 2021; Wen et al., 2021). Specifically, As levels increased by 94.8 ng m⁻³ and 82.1 ng m⁻³ at suburban and urban sites, respectively, while Zn levels increased by 37.3 ng m⁻³ and 48.9 ng m⁻³ at suburban and urban sites, respectively. However, there was a decrease in Pb
concentrations during the pollution period in both suburban sites (7.10 ng m$^{-3}$) and urban sites (8.52 ng m$^{-3}$). Pb serves as an important tracer for industrial processes, particularly in electroplating industry and mechanical manufacturing (Li et al., 2023). Additionally, changes in fuel composition, emissions control measures, and atmospheric conditions may also have contributed to the observed decrease in Pb concentrations (Li et al., 2023; Luo et al., 2021). Therefore, further source analysis is necessary.

3.2 Evaluation of Heavy Metals Pollution in PM$_{2.5}$

According to the pollution level of heavy metals, the I$_{geo}$ can be divided into seven categories ranging from uncontaminated to extremely contaminated (Table S2). Fig. 4(a) shows the I$_{geo}$ accumulation of heavy metals in clean period, pollution period, suburban site and urban site, respectively. Ba, Co, and Ti were at the “uncontaminated” (UC) category (I$_{geo}$ < 0), and Cr, Mn were within the “uncontaminated to moderately contaminated” (UC to MC) category (0 < I$_{geo}$ < 1) during the sampling period. This suggests a relatively lower influence of anthropogenic activity on these metals. In addition, the I$_{geo}$ values for As, Cu, Pb, Se, and Zn accounted for 56%–62% of
the accumulation fraction of total hazardous metals. The average $I_{geo}$ values of Cu, Pb, and Se exhibited varying degrees of decline during the pollution period, as depicted in Fig. 4(b), which was generally consistent with the observed trend in concentration changes. During the clean period, As was heavily contaminated, while during the pollution period, it was extremely contaminated, indicating that As was strongly influenced by anthropogenic activity. The comparison of $I_{geo}$ values for heavy metals between suburban and urban sites revealed similar levels of contamination (Fig. 4(c)). The mean values of $I_{geo}$ indicated that Cu exhibits moderate to heavy contamination, while Pb and Zn display heavy contamination. Se is found to be extremely contaminated throughout the observation period, providing further evidence of significant anthropogenic influences on these metals originating from sources such as combustion, traffic emission, and industrial manufacturing (Lao et al., 2022; Lin et al., 2020). Therefore, it is imperative to enhance the management of industrial activities and prioritize addressing the issue of heavy metal pollution in PM$_{2.5}$.

3.3 Potential Source Analysis

3.3.1 Source distribution for suburban and urban area

The source distribution between suburban and urban sites was employed by U.S. EPA PMF5.0 model. After conducting iterative testing on 4–10 factors in modelling exercises, the pattern of 5 factors was ultimately confirmed through performing 100 bootstrap runs, demonstrating relatively independent and comprehensive indicator sources. The contribution of 16 chemical components to the distribution of PM$_{2.5}$ concentration is shown in Text S5 and Fig. S3.

Fig. 5(a) shows the comparisons of pollution sources between suburban and urban sites. The contribution from mixed source (S1), crustal dust (S2), industry source (S3), vehicle exhaust (S4), and combustion sources (S5) were 27.7%, 16.1%, 13.2%, 27.7%, and 15.2% respectively in suburban sites, and 21.3%, 28.6%, 15.1%, 26.0%, and 9.0% respectively in urban sites. Vehicle exhaust account for a relatively high proportion in both urban and suburban areas. The proportion of combustion source in suburban areas was higher than that in urban areas, which might be associated with more severe open burning in the southern suburbs. In addition to the burning of garbage and straw, open-air barbecues are also an important source, related to the habits of the natives (Song et al., 2018). Given the absence of heating season in the study area, the proportion of combustion source is relatively lower compared to that observed in northern cities of China (Tian et al., 2018; Wu et al., 2020a). During the sampling period, there were several buildings under construction around the urban site, and earthworks caused a high proportion of dust.
The predominant industrial structures in the studied area include the metal smelting and processing industry, the food processing industry, and other manufacturing industries (Liu et al., 2019). This relatively singular industrial structure further confirms that the difference in source structure between suburban and urban areas are primarily attributed to the influence of human activities.

### 3.3.2 Source distribution for clean and pollution period

The PCA model was employed to examine the source distribution of PM$_{2.5}$ between clean and pollution period. The results of Kaiser-Meyer-Olkin (KMO) and Bartlett’s tests indicated that the data was applicable to PCA analysis, with KMO = 0.777 ($P = 0.000$) for clean period and KMO = 0.671 ($P = 0.000$) for the polluted period. Five principal components were extracted after varimax rotation PCA analysis, and the cumulative rate of contribution exceeded 90% (91.2% for clean period and 95.4% for pollution period), indicating the most information in PM$_{2.5}$ can be explained (Table S3 and Text S6).

The contribution of identified sources to PM$_{2.5}$ was further quantified through MLR analysis. The source contributions of PM$_{2.5}$ in clean period and pollution periods are presented in Fig. 5(b). During the clean period, mixed source ($\beta_1$), crustal dust ($\beta_2$), industry source ($\beta_3$), vehicle exhaust ($\beta_4$), and combustion sources ($\beta_5$) contributed 22.9%, 9.0%, 21.1%, 16.6%, and 30.4% respectively in the clean period, and 4.2%, 20.3%, 17.6%, 36.0%, and 22.0% respectively in the pollution period. The air quality in polluted days was primarily influenced by vehicle emissions, and crustal dust. Specifically, as pollution intensified, the contribution of crustal dust and vehicle exhaust increased by 11.3% and 19.4%, respectively. This result aligns with the observed increase in coarse particle concentrations during the pollution period. In addition, lower temperatures on polluted days led to increased vehicular activity, potentially contributing to a higher proportion of vehicle exhaust.

### 3.3.3 Source regions through the PSCF analysis

The HYPLIT model was employed to determine backward trajectories at 4-hour intervals. The 72-hour back trajectory with cluster calculation during the observation period is illustrated in Fig. 6(a). The air mass trajectory is divided into 3 categories. The type I flow from Jiangxi Province through Guangdong Province accounts for 67.4%, making it the main transport route. The type II airflow trajectory, passing through the junction of Hubei, Hunan, and Jiangxi Province, accounts for 13.6%. However, despite its relatively long transport distance, high-concentration pollution masses from northern regions can rapidly transit through this channel during the observation seasons under high-pressure conditions, thereby facilitating the transportation of pollutants to the study area (Zong et al., 2018). Additionally, the type III air masses transported by the sea through the coast of Guangdong accounted for 19.0%, bringing a large amounts of sea salt particles.
The PSCF analyses were conducted to further investigate the impact of regional emission sources on the variability of PM$_{2.5}$. The study area encompassed the geographical coordinates of 15°–35°N, 105°–125°E, with a spatial resolution of 0.2° × 0.2° in latitude and longitude. The starting elevation was established at 500 m above ground level. The PSCF map in Fig. 6(b) was generated based on hourly PM$_{2.5}$ concentrations. The largest WPSCF values were concentrated at the junction of Guangdong and Guangxi Provinces, indicating that this region had a strong source of pollution and was a potential source of PM$_{2.5}$. The WPSCF values showed a notable elevation for air masses transported through the western part of Jiangxi Province and the southeastern part of Hunan Province, influenced by northeast winds, indicating that emission sources in this region significantly contributed to air pollution in the studied area. Additionally, potential contributions were identified from the coastal areas of Guangdong Province. The influence of southeast winds facilitates the easy transport of pollutants over short distances from the coast of Guangdong Province and adjacent maritime areas to the studied region. In summary, the potential sources of PM$_{2.5}$ in the studied area are mainly associated with local pollution between neighboring cities and transmission from offshore areas. Therefore, it is imperative to enhance inter-regional collaboration in preventing and controlling pollution sources.

3.4 Human Health Risk Assessment

3.4.1 Concentration-based human health assessment

For carcinogenic risk, a range of $1 \times 10^{-6} < \text{CR} < 1 \times 10^{-4}$ indicates an acceptable level of carcinogenic risk for human, while CR $> 1 \times 10^{-4}$ suggests a cautionary level of carcinogenic risk for human. Regarding non-carcinogenic risk, the HQ $> 1$ signifies a non-carcinogenic risk for the population (Huang et al., 2021; Wang et al., 2022). The inhalation exposure results are presented in Fig. 7, illustrating the CR and HQ for each element. The carcinogenic risks were acceptable for all populations in each scenario (Fig. 7(a)). Cr and As dominated in total CRs for both adults and children. The CR associated with Co, Ni and Pb was consistently found to be less than $1 \times 10^{-6}$, indicating a negligible potential for carcinogenic risk. In comparison to the clean period, the CR values present an increasing trend during the pollution period due to an overall increase in concentration levels of TMs. Among them, As and Cr showed the largest increase in CR values.
As increased by $5.32 \times 10^{-5}$ in adults and $1.59 \times 10^{-5}$ in children. Cr increased by $1.35 \times 10^{-5}$ in adults, and $4.06 \times 10^{-6}$ in children. It is of concern that the CR values of As ($7.87 \times 10^{-5}$) for adults closely approach $1 \times 10^{-4}$, indicating a potential carcinogenic risk in the presence of more severe air pollution. In addition, the CR values at urban sites surpass those calculated at suburban sites.

The non-carcinogenic risks, as indicated by the HI values, were 2.28, 6.06, 2.73, and 3.18 for the clean period, pollution period, suburban sites, and urban sites, respectively, all of which exceeded the U.S. EPA standards (HI < 1), indicating a potential non-carcinogenic risk associated with all senarios (Fig. 7(d)). As, Mn, and V dominated the HI in each scenario. The highest HI was in pollution period, with HQ values ranked in decreasing order as follows: As (4.27) > V (1.09) > Mn (0.36).

Moreover, during the pollution period, both As and V showed an increase of over three times in their HQ values due to elevated concentrations of PM$_{2.5}$ along with enhanced levels of As and V content.

3.4.2 Source-based human health assessment

To evaluate the potential risks to human health associated with various sources of TMs, the contribution rates for CR and HI were calculated based on the results from source apportionment analysis. Different pollution sources contribute similarly to health risks in both children and adult (Huang et al., 2021; Jia et al., 2022; Wang et al., 2022). Therefore, the present study aims on compare the contribution rates of pollution sources during clean and polluted periods, as well as in urban and suburban areas.

As shown in Fig. 7(b), the comparison of source-based contribution to carcinogenic risk during clean and pollution periods revealed that combustion sources accounted for a significantly higher proportion (37.7%–45.0%) than other sources, which was also observed in non-carcinogenic risk fractions (Fig. 7(e)). This can be attributed to the fact that emissions from combustion sources ($β$5) contain more heavy metals both in clean and polluted period (Fig. S4). In addition, industrial sources presented higher health risks during the clean period, with a proportion of 28.2% for carcinogenic risk and 30.6% for non-carcinogenic risk. This is attributed to the industrial sources ($β$3) contributed more Cr (68.0%) and Ni (62.6%) during this period (Fig. S4(b)). It should be noted that mixed source posed a higher proportion of exposure risks during the pollution period. The contribution rate for carcinogenic risk increased by 12.7%, and for non-carcinogenic risk increased by 20.1% compared to the clean period.
The contribution fractions of the five sources to carcinogenic risk were found to be comparable between suburban and urban sites (Fig. 7(c)). Mixed source and industry source made the most significant contribution to the carcinogenic risk values throughout the observation period, accounting for 26.1%–32.4% and 29.2%–31.6%, respectively. The health risks associated with combustion sources were higher in suburban sites compared to urban sites, exhibiting an increase of 7.4% and 1.0% for carcinogenic and non-carcinogenic risks, respectively. The contribution of industry sources to carcinogenic risk (Fig. 7(c)) was significantly higher compared to non-carcinogenic risks (Fig. 7(f)), both in suburban and urban sites. Therefore, greater emphasis should be placed on assessing the impact of industrial activities on the population’s exposure to carcinogenic risk. The health risks associated with crustal dust were found to be significantly higher in urban areas compared to suburban areas, both in terms of carcinogenic and non-carcinogenic effects. This can be attributed to the elevated levels of heavy metals present in road dust within urban environments. To mitigate health risks to the population, the government should focus on policies aimed at preventing and controlling atmospheric heavy metal pollution, while enhancing integrated management of multiple pollution sources.

4 CONCLUSIONS

The pollution levels in urban areas typically exhibited higher concentrations compared to those in suburban areas. The concentrations of PM$_{2.5}$ were $90.8 \pm 13.3 \mu g m^{-3}$ and $101.8 \pm 21.0 \mu g m^{-3}$ in suburban and urban sites, respectively, during the polluted period. SNA were the primary contributor to PM$_{2.5}$ concentration. The air quality on polluted days were mainly affected by vehicle exhaust (36.0%), combustion sources (22.0%), and crustal dust (20.3%). Moreover, the pollution episode was characterized by a significant increase in coarse particle concentration. The potential sources of PM$_{2.5}$ in the studied area were identified as local pollution originating from neighboring cities and offshore transmission.

The distribution of the geo-accumulation index highlighted significant anthropogenic influences on the accumulation of trace metals. Specifically, As, Cu, Pb, Se, and Zn were identified as priority harmful species. Cr and As emerged as the primary contributors to the overall carcinogenic risk in both adults and children. The non-carcinogenic risks cannot be ignored in each scenario, especially for As and V. The combustion sources were the most important factor causing carcinogenic risk and non-carcinogenic risk both in clean and pollution period. Crustal dust in urban sites posed greater health risks compared to suburban sites, encompassing both carcinogenic and non-carcinogenic human health risks. The present study provides scientific evidence to support the establishment of a comprehensive database focusing on spatiotemporal variability and source apportionment of PM$_{2.5}$ in underdeveloped regions. Furthermore, it is crucial to emphasize the integrated management of multiple pollution sources.

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SUPPLEMENTARY MATERIAL

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