

Article

Temporal Variations, Source Attributions, and Health Risks of PM_{2.5}-Bound Trace Elements in a Megacity in Central China

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Abstract

Trace elements bound to PM_{2.5} constitute crucial factors increasing human health risks. To optimize health-oriented air quality management strategies for industrial cities, this study monitored major trace elements in PM_{2.5} in Wuhan, China, from June 2023 to May 2024. During the study period, the mean concentration of trace elements was 3290 ± 2270 ng/m³, comprising 93.9% crustal elements and 6.1% heavy metals. The positive matrix factorization model identified fugitive dust as the primary source of trace elements, with a mean contribution of 35.6%, followed by traffic emissions (21.8%), industrial emissions (19.1%), biomass and waste incineration (12.3%), and coal combustion (11.1%). Health risk assessment revealed that the cumulative non-carcinogenic and carcinogenic risks of trace elements via inhalation exposure exceeded acceptable levels for both local children and adults, with Mn, As, and Cr as key risk factors. Although coal combustion contributed less to trace element concentrations than other sources, it was the largest contributor to non-carcinogenic and carcinogenic risks, followed by fugitive dust and traffic emissions. These results suggest that trace element pollution control in Wuhan should prioritize reducing coal combustion emissions, while emphasizing the critical importance of sustained traffic regulation and fugitive dust suppression to improve air quality and protect public health.

Keywords: industrial urban pollution; crustal elements; heavy metals; air pollution mitigation



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1. Introduction

PM_{2.5} has long been a globally concerning air pollutant due to its well-documented adverse impacts on air quality, human health, and climate change [1,2]. Trace elements are the primary components contributing to PM_{2.5} toxicity, capable of being directly inhaled into the human respiratory tract alongside PM_{2.5}, penetrating deep into alveoli and entering the bloodstream [3]. Toxicological and epidemiological studies have demonstrated that trace elements exert toxic effects on human organs and tissues through mechanisms such as inducing abnormal DNA methylation and oxidative damage [4]. For instance, exposure to Fe in PM_{2.5} induces hematological disorders and increased anemia risk; Cu

has been associated with elevated dementia incidence; Ca, Fe, Mn, and Pb are implicated in exacerbating asthma; and exposure to As and Cr can lead to skin lesions, neurological symptoms, respiratory diseases, cardiovascular disorders, and cancer [5–9].

Trace elements originate from natural processes, such as floating soil dust and sandstorms, and anthropogenic activities, including traffic emissions, coal combustion, biomass burning, waste incineration, metal smelting, and steel production [10,11]. Owing to their source-indicative properties, trace elements are widely used to identify PM_{2.5} sources. Numerous studies have employed the Positive Matrix Factorization (PMF) model to determine the sources of PM_{2.5}-bound trace elements across various cities and regions [12–14]. For example, shipping emissions are the primary source in coastal areas, while sand and soil dust constitute the largest contributor in arid areas [13–15]. Quantifying source contributions enables source-oriented health risk assessment, thereby improving the accuracy and effectiveness of trace metal pollution control strategies [16,17].

Industrial cities are hotspots for trace element emissions, with their complex urban environments introducing high uncertainties in pollution characteristics, source compositions, and associated health risks [18,19]. As a major industrial base and transportation hub in Central China, Wuhan hosts a pillar industrial cluster centered on steel production, automotive manufacturing, and petrochemicals [20]. In recent years, rapid industrialization and urbanization have posed persistent challenges to air quality [3,21]. Despite government-implemented air pollution control measures, PM_{2.5} concentrations in Wuhan remain chronically above the corresponding limits set by the Ministry of Ecology and Environment of China and the World Health Organization (WHO) [22,23]. Previous studies on PM_{2.5} pollution in Wuhan have primarily focused on spatiotemporal distributions and meteorological influences, with limited attention to compositional characteristics of PM_{2.5}, particularly trace elements [15,21,24,25]. This knowledge gap hinders the precise identification of key PM_{2.5} contributors and high-health-risk pollution sources, which is critical for formulating emission reduction strategies and public health policies.

This study conducted PM_{2.5} sampling in Wuhan from June 2023 to May 2024, analyzing temporal variations in the concentrations of PM_{2.5}-bound trace elements, quantifying sources via PMF modeling, and evaluating non-carcinogenic and carcinogenic risks of trace elements and their sources to residents. The findings aim to deepen understanding of trace element pollution during local air pollution control processes and provide scientific guidance for refining health-oriented air quality management strategies in Wuhan and other industrial cities.

2. Materials and Methods

2.1. Sampling Design

PM_{2.5} samples were collected on the rooftop of the Resources and Environment Building at Hubei University of Technology, Wuhan, China (30.48° N, 114.31° E; Figure 1), at an elevation of approximately 10 m above ground level, using a high-volume air sampler (TH-1000CII, Wuhan Tianhong Co., Ltd., Wuhan, China) paired with quartz fiber filters (8 × 10 inch, 2500QAT-UP, Pallflex Products, Putnam, CT, USA) operating at a constant flow rate of 1.0 m³/min. The sampling site is situated 300 m west of a main road and approximately 300 m south of the Yangsigang Expressway, which carries an average daily traffic volume of 58,000 vehicles (Wuhan Transportation Bureau, <https://jtj.wuhan.gov.cn/>, accessed on 22 June 2026). A waste incineration plant is located 11.6 km south of the site, and multiple construction sites for buildings and roads were ongoing within a 3 km radius during the sampling period. The sampling site lies in central Wuhan, surrounded by integrated cultural, residential, and commercial zones and major arterial roads, which well

represent urban atmospheric pollution features of downtown Wuhan. Detailed information is provided in Text S1 of the Supplementary Materials.

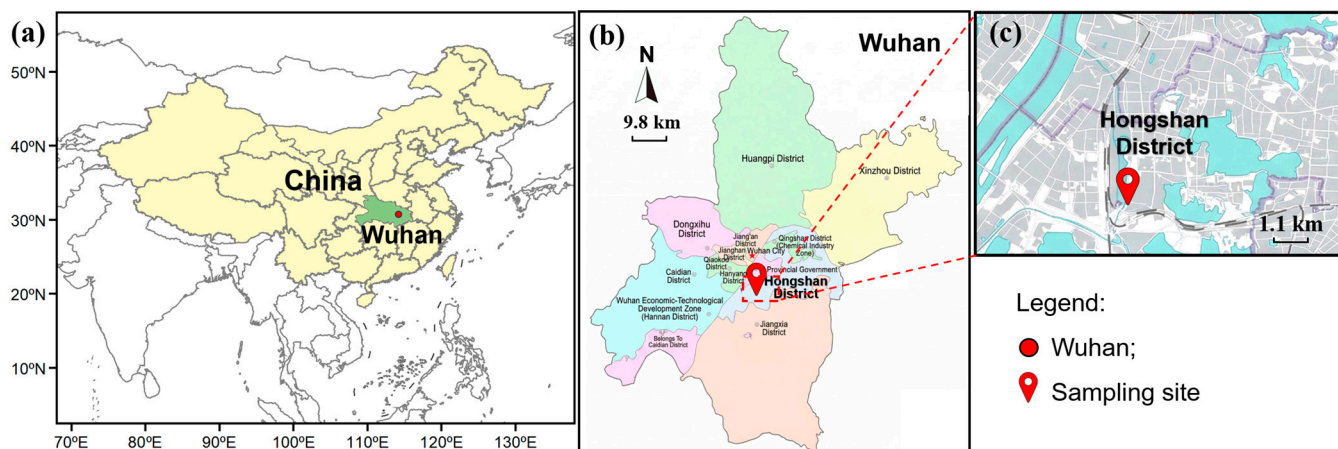


Figure 1. Locations of (a) Wuhan in China, (b) the sampling site in Wuhan, and (c) the sampling site in Hongshan District.

Sampling was conducted from June 2023 to May 2024, encompassing four seasons: spring (March–May), summer (June–July), autumn (September–November), and winter (December–February). Specific sampling dates are detailed in Table S1. Except for the winter months, continuous sampling was performed for 14 days each month. Given that air pollution typically exacerbates in winter, intensive sampling was conducted in December and January, with durations of 17 and 25 days, respectively; however, sampling in February was interrupted due to freezing rain events. Daytime and nighttime samples were collected on each sampling day from 08:00 to 18:00 and from 20:00 to 06:00 the subsequent morning, respectively. A total of 336 PM_{2.5} samples and 11 field blank filters were obtained. Analytical measurements of the blank filters confirmed the absence of target trace elements, indicating that contamination throughout the sampling process was negligible. Hourly meteorological parameters, including temperature (T), relative humidity (RH), wind speed (WS), rainfall (RF), and pressure (P), in Wuhan during the sampling period were obtained from the National Meteorological Center of China (<http://www.nmc.cn/publish/forecast/AHB/wuhan.html>, accessed on 22 June 2026). Boundary layer height (BLH) was acquired from the European Centre for Medium-Range Weather Forecasts ERA5 reanalysis [26].

2.2. Trace Elements Analyses

Twenty-one trace elements were detected in each PM_{2.5} sample, categorized as 6 crustal elements (Al, Fe, Ca, Na, K, Mg) and 15 heavy metals (Zn, Mn, Cu, Pb, Ba, Sr, As, Se, Cr, V, Ni, Co, Bi, Cd, Ga). Standards for crustal element quantification and the internal standard, an indium standard solution (1000 mg/L in HNO₃), were purchased from Wako Pure Chemicals (Osaka, Japan). The XSTC-13 mixed standard for heavy metal quantification was provided by SPEX CertiPrep (Metuchen, NJ, USA). All reagents utilized in the analyses were of analytical reagent grade.

Sample pretreatment and elemental analytical procedures were consistent with those described in previous studies [27,28]. Briefly, filter fragments were extracted three times using dilute nitric acid via ultrasound heating, followed by dark equilibration and membrane filtration before instrumental analysis. Crustal elements and heavy metals were determined using inductively coupled plasma optical emission spectroscopy (ICP-OES, ES-710, Varian, Inc., Palo Alto, CA, USA) and inductively coupled plasma mass spectrometry (ICP-MS, 7800, Agilent, Santa Clara, CA, USA).

try (ICP-MS, iCAP RQ, Thermo Fisher Scientific Corp., Waltham, MA, USA), respectively. Calibration curves showed satisfactory linearity, and internal standard correction guaranteed acceptable analyte recoveries between 90% and 110%. All method detection limits are summarized in Table S2. Detailed experimental operations and quality assurance/quality control procedures are provided in Texts S1 and S2 in the Supplementary Materials.

2.3. Source Apportionment

In this study, the PMF (version 5.0) model developed by the U.S. Environmental Protection Agency (USEPA) was employed to quantitatively identify the sources of trace elements. Input data for the PMF model comprised the mass concentrations and corresponding uncertainties of 21 trace elements across all samples. Uncertainties were calculated using the method detection limits and measurement error fraction [29]. PMF solutions with 3 to 7 factors were evaluated in this study, and the optimal 5-factor solution was ultimately selected based on interpretability and error estimation results. Bootstrap (BS), displacement (DISP), and BS-DISP approaches were adopted for error estimation to guarantee the robustness of PMF solutions. The methods for calculating the uncertainty and determining the optimal factor solution are presented in Text S3 and Tables S3 and S4.

2.4. Backwards Trajectory Clustering

During the study period, the main pathways of airflow trajectories reaching the sampling site were calculated using the Hybrid Single-Particle Lagrangian Integrated Trajectory (HYSPLIT) model with meteorological data from the Global Data Assimilation System ($1^\circ \times 1^\circ$ spatial resolution). Synchronized with the daily day-night sampling times, 72 h backward trajectories were computed hourly from the sampling site at 500 m AGL. Then, a monthly clustering analysis was performed on the obtained trajectories using the clustering module within the HYSPLIT model. Model operation was consistent with that described in the previous study [30].

2.5. Health Risk Estimation

The carcinogenic and non-carcinogenic health risks posed by PM_{2.5}-bound trace elements via inhalation exposure to local children and adults were estimated using a modified USEPA health risk assessment model [16,31]. First, the exposure concentration (EC) was calculated using Equation (1). On this basis, the non-carcinogenic risk (hazard quotient, HQ) was calculated using Equations (2) and (3) for 9 non-carcinogenic elements (Ni, As, Zn, Cu, Co, Cr, V, Mn, Pb, Ba). The carcinogenic risk (CR) was calculated using Equations (4) and (5) for 5 carcinogenic elements (Ni, As, Co, Cr, Pb).

$$EC = C \times IR \times ET \times EF \times ED / (BW \times AT) \quad (1)$$

where C represents the concentration of a trace element in PM_{2.5}, $\mu\text{g}/\text{m}^3$; IR denotes the inhalation rate for adults ($1.5 \text{ m}^3/\text{h}$) and children ($0.3625 \text{ m}^3/\text{h}$); ET denotes daily exposure time, $24 \text{ h}/\text{d}$; EF denotes exposure frequency, $350 \text{ d}/\text{a}$; ED denotes the exposure duration, 6 years for children and 24 years for adults; BW denotes the body weight for adults (73 kg) and children (25.5 kg); and AT is the averaging time, defined as $ED \times 365 \text{ d}/\text{a} \times 24 \text{ h}/\text{d}$ for non-carcinogens and $70 \text{ years} \times 365 \text{ d}/\text{a} \times 24 \text{ h}/\text{d}$ for carcinogens.

$$HQ = EC / RfD \times 1000 \mu\text{g}/\text{mg} \quad (2)$$

$$RfD = RfC \times IR / BW \quad (3)$$

where RfC is the chronic inhalation reference concentration, mg/m^3 ; RfD is the reference dose, $\text{mg}/(\text{kg} \cdot \text{day})$. The cumulative non-carcinogenic risk of multiple trace elements was

estimated using the hazard index (*HI*), which is the sum of *HQ* values for individual elements. An *HQ* or *HI* value of 1 or less indicates a low probability of non-carcinogenic effects, whereas a value exceeding 1 suggests a high probability of non-carcinogenic effects [32].

$$CR = EC \times CSF \quad (4)$$

$$CSF = IUR \times BW / IR \quad (5)$$

where *IUR* represents the inhalation unit risk, $(\mu\text{g}/\text{m}^3)^{-1}$; *CSF* is the cancer slope factor, $(\text{kg} \cdot \text{day})/\text{mg}$. The concentration of Cr(VI) for *CR* calculation is considered as 1/7 of the total Cr concentration [33]. The USEPA recommends a tolerable *CR* range for regulatory purposes of 10^{-6} to 10^{-4} [31]. A *CR* value below 10^{-6} indicates no significant carcinogenic risk, while a *CR* value above 10^{-4} suggests a serious carcinogenic risk [16]. The *RfC* and *IUR* values (Table S2) used in this study were obtained from USEPA regional screening levels [32].

Furthermore, based on the output of the PMF model, the health risks associated with each source of trace metals were estimated [13]. The concentration of each element derived from different sources was calculated using Equation (6). Subsequently, the *HQ* and *CR* for each source were estimated through Equations (1)–(5).

$$C_{ij}^k = g_{ik} \times f_{kj} \quad (6)$$

where C_{ij}^k represents the mass concentration of the *j*-th element from the *k*-th source in the *i*-th sample, $\mu\text{g}/\text{m}^3$; g_{ik} denotes the relative contribution of the *k*-th source to the *i*-th sample, %; and f_{kj} is the concentration of the *j*-th element from the *k*-th source, $\mu\text{g}/\text{m}^3$. Both g_{ik} and f_{kj} are outputs from the PMF model.

3. Results and Discussion

3.1. Overview of Trace Element Concentrations

Table 1 presents the mean concentrations and standard deviations of trace elements in PM_{2.5} during the sampling period. The mean concentration of total trace elements was $3290 \pm 2270 \text{ ng}/\text{m}^3$, dominated by crustal elements ($3090 \pm 2230 \text{ ng}/\text{m}^3$), accounting for 93.9% of the total. The most abundant crustal element was Ca ($1520 \pm 1240 \text{ ng}/\text{m}^3$), followed by K ($513 \pm 342 \text{ ng}/\text{m}^3$) and Fe ($382 \pm 261 \text{ ng}/\text{m}^3$), while concentrations of Al ($249 \pm 217 \text{ ng}/\text{m}^3$), Mg ($217 \pm 188 \text{ ng}/\text{m}^3$), and Na ($207 \pm 151 \text{ ng}/\text{m}^3$) were comparatively lower. The mean concentration of heavy metals was $197 \pm 66.0 \text{ ng}/\text{m}^3$, with primary components including Zn ($84.2 \pm 42.6 \text{ ng}/\text{m}^3$), Mn ($30.0 \pm 15.9 \text{ ng}/\text{m}^3$), Pb ($23.9 \pm 14.8 \text{ ng}/\text{m}^3$), Ba ($17.9 \pm 10.9 \text{ ng}/\text{m}^3$), and Cu ($13.4 \pm 7.35 \text{ ng}/\text{m}^3$), collectively accounting for 85.6% of total heavy metals. The compositional characteristics of high Zn, Pb, Cu, and Mn contents align with those observed in most cities across China, which are closely associated with industrial emissions and fossil fuel combustion [15,34].

Compared with previous PM_{2.5} observations in Wuhan (Table S5), concentrations of the same trace elements except Ca were reduced in this study and fell below the average levels for southwestern and central China in 2013, reflecting the positive effects of Wuhan's comprehensive air pollution control measures in recent years [3,11,15,19,35–43]. Moreover, the concentrations of most trace elements observed in this study were comparable in magnitude to those in other Chinese cities (Table S5), while the Ca concentration was notably elevated, potentially attributable to extensive construction activity in Wuhan.

Table 1. Overall and seasonal mean concentrations and standard deviations (ng/m³) of trace elements in PM_{2.5} in Wuhan during the sampling period.

Species	Overall	Spring	Summer	Autumn	Winter
Trace elements	3290 ± 2270	4180 ± 2830	2200 ± 751	3080 ± 2910	3900 ± 1100
Crustal elements	3090 ± 2230	3950 ± 2790	2070 ± 69.5	2890 ± 2750	3630 ± 1080
Al	249 ± 217	343 ± 296	181 ± 71.9	221 ± 192	255 ± 88.5
Fe	382 ± 261	472 ± 312	253 ± 80.2	378 ± 316	446 ± 163
Ca	1520 ± 1240	2050 ± 1770	1060 ± 370	1430 ± 1270	1570 ± 776
Na	207 ± 151	235 ± 186	152 ± 99.6	173 ± 146	300 ± 91.2
K	513 ± 342	620 ± 322	261 ± 133	456 ± 238	819 ± 358
Mg	217 ± 188	239 ± 197	170 ± 89.7	227 ± 192	241 ± 90.4
Heavy metals	197 ± 66.0	227 ± 33.4	124 ± 90.6	193 ± 99.2	266 ± 92.4
Zn	84.2 ± 42.6	94.5 ± 26.9	51.5 ± 17.9	90.4 ± 44.6	108 ± 50.5
Mn	30 ± 15.9	35 ± 18.3	20.2 ± 6.62	293 ± 16.5	37.7 ± 13.5
Cu	13.4 ± 7.35	15.4 ± 6.25	9.1 ± 2.56	12.2 ± 6.97	18.7 ± 8.38
Pb	23.9 ± 14.8	27.4 ± 11.3	14.8 ± 4.58	22.1 ± 14.2	35 ± 17.3
Ba	17.9 ± 10.9	22.5 ± 10.7	9.15 ± 2.6	14.3 ± 8.94	29.8 ± 10.4
Sr	8.64 ± 7.42	10.8 ± 8.93	5.81 ± 4.61	7.66 ± 6.07	11.1 ± 6.01
As	6.29 ± 5.28	8.61 ± 5.58	3.87 ± 2.16	5.36 ± 4.82	7.83 ± 6.18
Se	3.67 ± 1.75	3.81 ± 1.31	3.64 ± 1.62	3.34 ± 1.62	4 ± 2.28
Cr	2.69 ± 1.21	2.44 ± 0.88	2.64 ± 1.19	2.6 ± 1.2	3.28 ± 1.38
V	0.88 ± 0.53	1.27 ± 0.66	0.63 ± 0.24	0.75 ± 0.58	0.88 ± 0.27
Ni	0.83 ± 0.53	1.09 ± 0.56	0.65 ± 0.38	0.74 ± 0.53	0.85 ± 0.39
Co	0.23 ± 0.19	0.35 ± 0.25	0.12 ± 0.05	0.21 ± 0.19	0.23 ± 0.09
Rb	1.3 ± 0.77	1.7 ± 0.98	0.76 ± 0.25	1.19 ± 0.58	1.7 ± 0.64
Bi	2.49 ± 1.71	2.31 ± 1.43	1.22 ± 0.77	1.69 ± 1.05	5.87 ± 4.85
Cd	1.56 ± 1.03	1.61 ± 0.79	0.95 ± 0.67	1.8 ± 1.32	2.04 ± 1.44
Ga	0.33 ± 0.21	0.34 ± 0.14	0.2 ± 0.08	0.29 ± 0.12	0.56 ± 0.25
Cs	0.12 ± 0.07	0.14 ± 0.08	0.08 ± 0.03	0.12 ± 0.07	0.13 ± 0.06

Based on the air quality guidelines stipulated by the Ministry of Ecology and Environment of China and the WHO (Table 2), the mean concentrations of Mn, Pb, and V in this study were far below their respective guideline values [22,44]. However, the mean As concentration (6.29 ± 5.28 ng/m³) exceeded the limit of China (6 ng/m³), and the mean Cr concentration (2.69 ± 1.21 ng/m³) exceeded both the guideline values for Cr(VI) (accounting for 1/7 of the total Cr concentration) of China (0.025 ng/m³) and WHO (0.25 ng/m³) [22,33,44]. High concentrations of As and Cr in PM_{2.5} have also been observed recently in many Chinese cities (Table S5), such as Hangzhou (As: 95 ng/m³; Cr: 41 ng/m³), Linfen (56 ng/m³; 3.3 ng/m³), Nanjing (12 ng/m³; 6 ng/m³), Beijing (11 ng/m³; 3.6 ng/m³), and Zhengzhou (8 ng/m³; 4 ng/m³), indicating that their exceedances have become a prevalent air pollution issue across China, posing a threat to public health [18,38–41]. Although vigorous implementation of air pollution control measures in China has effectively reduced the concentrations of toxic elements like As and Cr in urban PM_{2.5}, the above findings suggest that Chinese authorities should further mitigate trace element pollution to prevent sustained adverse impacts on public health [45,46].

Table 2. Guideline values for trace element concentrations in ambient air stipulated by the Ministry of Ecology and Environment of China and the WHO.

Species	China Guideline Values (ng/m ³) ^a	WHO Guideline Values (ng/m ³) ^b
Mn	n.a. ^c	150
Pb	500	500
As	6	6.6
Cr(VI)	0.025	0.25
V	n.a. ^c	1000
Cd	5	5

^a Data cited from [22]. ^b Data cited from [44]. ^c Not available.

3.2. Temporal Variations in Trace Elements

3.2.1. Seasonal Trends

The concentrations of trace elements exhibited obvious seasonal variations during the sampling period (Figure 2). The seasonal trend for crustal elements followed spring > winter > autumn > summer, whereas heavy metals showed winter > spring > autumn > summer. Differences in the sources of crustal elements and heavy metals possibly account for their distinct seasonal patterns.

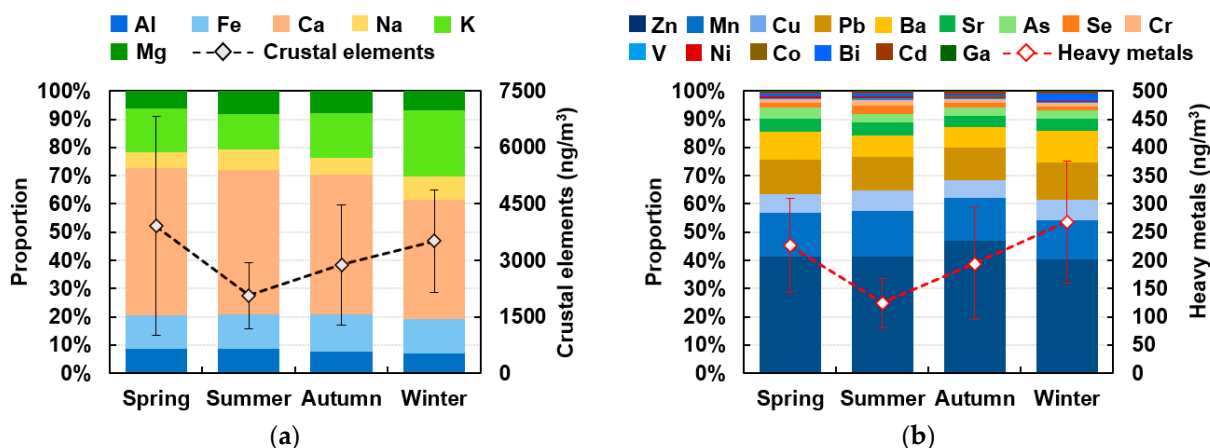


Figure 2. Seasonal variations in the proportion and concentration of (a) crustal elements and (b) heavy metals during the sampling period. Vertical lines extending from diamond markers represent standard deviation.

Crustal elements are primarily associated with fugitive dust emissions, such as resuspension of local soil and sand dust [47]. As shown in Table S6, Wuhan experienced lower RH in spring (67.4%) and winter (68.1%) than in summer (71.7%) and autumn (75.0%). Dry weather conditions intensified fugitive dust emissions, leading to increased suspension of dust particles in the ambient air [48]. Furthermore, large-scale dust storms prevail in northwestern China during spring, followed by winter [49]. Long-range transport of sand dust could exacerbate dust pollution in Wuhan, thereby increasing concentrations of crustal elements [50]. For instance, during a dust event in northern China from 19 to 20 March 2024, daily concentrations of crustal elements in Wuhan surged from 3690 ng/m³ on 18 March to 12,840 ng/m³ on 19 March, while the concentration of heavy metals showed no obvious variation (Figure S1a). Concurrently, concentrations of Ca, Al, Fe, Mg, and Na on 19 March increased by 2.6- to 4.8-fold compared to the previous day (Figure S1b), indicating a large contribution from sand dust to these elements. In contrast, dust storms rarely occur in summer and autumn [49]. Moreover, higher RH and RF during these seasons (Table S6) facilitated the wet deposition of dust particles, which alleviated dust pollution and reduced the concentrations of crustal elements. In addition, Ca displayed relatively elevated levels across all seasons (Figure 2), suggesting additional contributions from human disturbance, including construction and road traffic dust emissions [13]. This observation is consistent with the rapid urbanization of Wuhan: in 2023, the total construction area of buildings reached 144.6 million m², and vehicle ownership exceeded 4.46 million [51]. Beyond natural dust sources, K is a typical tracer of biomass burning [52]. Higher K concentrations were observed in autumn and winter, potentially related to enhanced biomass burning activities during these seasons.

Different from crustal elements, heavy metals primarily originate from anthropogenic emissions [48]. Wuhan hosts concentrated heavy industries, including automotive manufacturing, steel production, petrochemicals, and metallurgy, alongside extensive transportation

networks; therefore, emissions from industrial processes, traffic, and combustion activities have been recognized as the major anthropogenic sources of local air pollutants [20]. The seasonal pattern of heavy metal concentrations peaking in winter has been widely reported in existing studies [3,11]. Temperature inversions and stagnant atmospheric conditions are prevalent in winter, which compresses the BLH and inhibits the diffusion of air pollutants, leading to their local accumulation [53]. Moreover, pollutants emitted from extensive residential heating activities in northern China during winter could be transported southward by Siberian cold air masses and further exacerbate air pollution in Wuhan [54]. In other seasons, the BLH was higher than that in winter (Table S6), which facilitates the diffusion and dilution of air pollutants, thereby reducing the concentrations of heavy metals [53].

3.2.2. Weekly and Diurnal Variations

Figure 3 compares the concentrations of trace elements between weekdays and weekends. During the sampling period, the mean concentration of each trace element was higher on weekdays than on weekends. Similar intra-week patterns have been observed in previous studies conducted in Shanghai (China), Lisbon (Portugal), New Jersey (USA), and Agra (India) [55–58]. However, an inverse trend (higher concentrations on weekends) was reported for Beijing (China) [59].

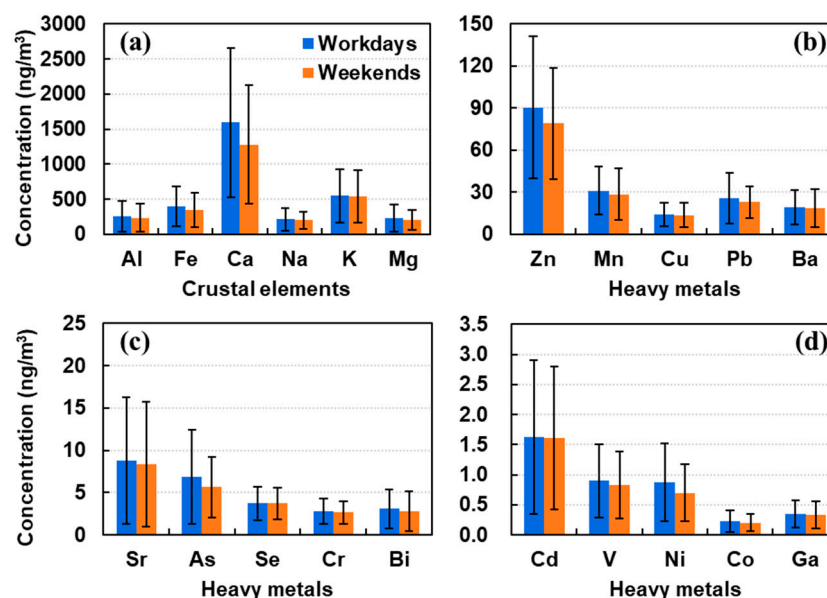


Figure 3. Differences in concentrations of (a) crustal elements and (b–d) heavy metals between weekdays and weekends during the sampling period. Black lines on bars denote standard deviation.

Energy-intensive industries usually operate year-round, requiring their core production processes to remain uninterrupted: for example, boilers in power plants require consecutive combustion, blast furnaces in steel mills must sustain high temperatures, and reactors in petrochemical plants operate continuously [19,39]. Consequently, the daily emission intensity of these industries remains stable. In comparison, traffic emission intensity exhibits pronounced intra-week variations, with traffic volumes generally higher on weekdays than on weekends [55,57,58]. Congestion during morning and evening rush hours on weekdays is particularly prevalent, leading to idling or low-speed driving and frequent braking, which further exacerbate traffic emissions and pollutant concentrations [60]. Notably, Beijing has implemented a vehicle restriction policy on weekdays since 2008, which has stimulated increasing weekend traffic volumes and emissions, leading to higher trace element concentrations on weekends than on weekdays [59]. Based on the correlation between trace element concentrations and traffic activity intensity, the

forementioned studies suggest that intra-week variations in trace element concentrations are dominated by traffic emissions. In the present study, obvious intra-week differences were observed in the concentrations of Ca, Zn, As, and Ni, with weekday/weekend ratios above 1.2 (Figure 3). These elements are associated with vehicle emissions and road dust, indicating that their elevated concentrations on weekdays were possibly attributed to heightened traffic activity [48,61].

The diurnal differences in trace element concentrations during the sampling period are described in Figure 4. Owing to more frequent human activities and disturbances during daytime, the emission intensities of fugitive dust sources and anthropogenic sources were enhanced, resulting in higher daytime concentrations of most trace elements compared to nighttime concentrations [39]. In contrast, the concentrations of K, Cu, Bi, and Ba were higher at night than during the day, with this diurnal difference being particularly pronounced in winter (Figure S2). As shown in the wind rose diagrams of the diurnal concentrations of these four elements (Figure S3a–d), wind speeds were generally higher during the day, and enhanced atmospheric dilution reduced the ambient concentrations of K, Cu, Bi, and Ba, whereas stable temperature inversion frequently formed at night, which trapped local pollutants near the ground, further increasing their concentrations [24]. Cu, Bi, and Ba are emitted from diverse sources, encompassing coal combustion, biomass burning, waste incineration, transportation, and industrial emissions [13,37]. High concentrations of K at night were mainly observed under northeasterly and southeasterly wind conditions (Figure S3a), suggesting that biomass burning intensified during the night, possibly originating from Wuhan and its surrounding areas. Meanwhile, low-speed southerly winds at night led to increased concentrations of Ba, Bi, and Cu (Figure S3b–d), which may be related to emissions from the waste incineration plant located approximately 11.6 km to the south of the sampling site. Based on their distinct diurnal distribution compared to characteristic elements from coal combustion, transportation, and industrial emissions (e.g., Zn, Pb, Ni), the elevation in concentrations of K, Cu, Bi, and Ba at night was likely related to biomass burning and waste incineration [18,61]. Collectively, these observations highlight the necessity of strengthened supervision and emission control over nocturnal pollution activities to comprehensively improve ambient air quality.

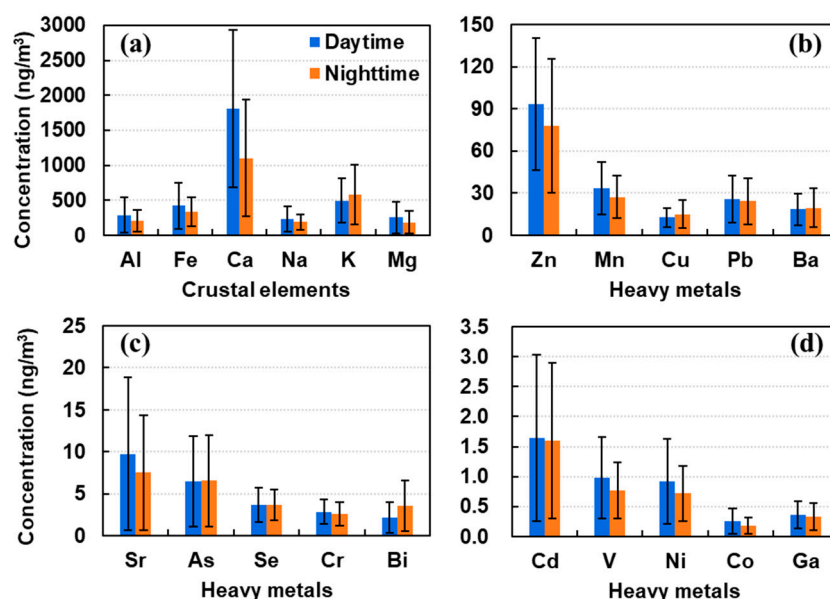


Figure 4. Differences in concentrations of (a) crustal elements and (b–d) heavy metals between daytime and nighttime during the sampling period. Black lines on bars denote standard deviation.

3.3. Main Sources of Trace Elements

The PMF model identified five source factors for PM_{2.5}-bound trace elements in this study; the factor profiles are shown in Figure S4. Factor 1 was characterized by a high content of Zn, accounting for 65% of the total factor contribution, followed by Ga, Mn, Pb, Se, and Fe, with each proportion exceeding 30%. Zn, Ga, and Fe serve as crucial raw materials in metal smelting, steel production, and alloy manufacturing, while Mn, Pb, and Se mainly originate from high-temperature processes such as roasting and sintering [62,63]. The high loading of these elements aligns with steel-based heavy industry in Wuhan. Petrochemical and steel industrial zones are located to the northeast of the sampling site, while manufacturing and automotive-related factories are situated in the east to southeast [3,25]. As shown in the wind rose diagram of factor contributions (Figure S5a), prevailing northeasterly and southeasterly winds increased the contribution of factor 1, further confirming that factor 1 corresponds to industrial emissions (IE). Factor 2 showed prominent proportions of As and Cd (>60%), with moderate proportions from Pb and Bi (approximately 40%). Since these elements are major components of coal combustion emissions, factor 2 was identified as coal combustion (CC) [18,54]. The wind rose diagram (Figure S5b) shows that factor 2 exhibited a similar directional-dependence pattern to factor 1, indicating that coal-derived pollutants were primarily emitted by industrial facilities. This observation is consistent with the fact that coal consumption in Wuhan is mainly driven by industrial activities [21], thereby verifying the identification of factor 2 as CC.

Factor 3 was dominated by Cr and Ni (>50%), with Se, V, and Cu accounting for more than 20%. Cr, Ni, Se, and V are tracers of petroleum combustion, while Cu serves as a catalyst material for purifying vehicle exhaust and constitutes brake pad and tire wear debris [11,64]. The sampling site is surrounded by busy roadways on its northern, southern, and eastern sides. Traffic-related emissions from these sides led to a relatively stable contribution of factor 3 with changes in wind direction (Figure S5c). Consequently, factor 3 was classified as traffic emissions (TE). Factor 4 primarily consisted of Ca, Mg, Al, Sr, Co, Fe, Na, V, and Ni, with each proportion above 50%. This composition is consistent with the characteristic profile of fugitive dust. The wind rose diagram for factor 4 shows a similar distribution to that of factor 3 (Figure S5c,d), indicating that road dust is the main component of factor 4. Moreover, pollutants carried by northeasterly winds substantially increased the contribution of factor 4, which might be associated with road and construction activities to the northeast of the sampling site. Based on these observations, factor 4 was attributed to fugitive dust (FD) [48,50].

Factor 5 exhibited a high proportion of K (50%), followed by Ba (45%), Bi (45%), and Cu (35%). As previously noted, K is a tracer for biomass burning, while Ba, Bi, and Cu can be emitted from biomass burning and waste incineration [52,65]. As illustrated in Section 3.2.2 and the wind rose diagrams (Figure S3a–d), elevated K concentrations under winds from northeast to southeast directions possibly originated from biomass burning activities in Wuhan and the surrounding areas. The distribution of high concentrations of Ba, Bi, and Cu partially overlapped with that of K, while the notable rise in these three elements under southerly winds can be attributed to emissions from the waste incineration plant situated to the south of the sampling site. In summary, Factor 5 represents the combined contribution of biomass burning and waste incineration (BWI).

Figure 5a presents the mean contribution of the five factors throughout the sampling period. FD was the dominant source of trace elements in Wuhan, with a mean contribution of 35.6% over the entire sampling period. This result is consistent with the high contributions of FD sources (ranging from 21.4% to 42.7%) reported in northern, eastern, and coastal cities in China, primarily attributable to construction activities, road dust resuspension, and sand dust events [13,40]. TE, IE, BWI, and CC constituted the principal anthropogenic

sources of trace elements, with mean contributions of 21.8%, 19.1%, 12.3%, and 11.1%, respectively. This source composition aligns with the characteristics of developed transport infrastructure and intensive heavy industry in Wuhan [3].

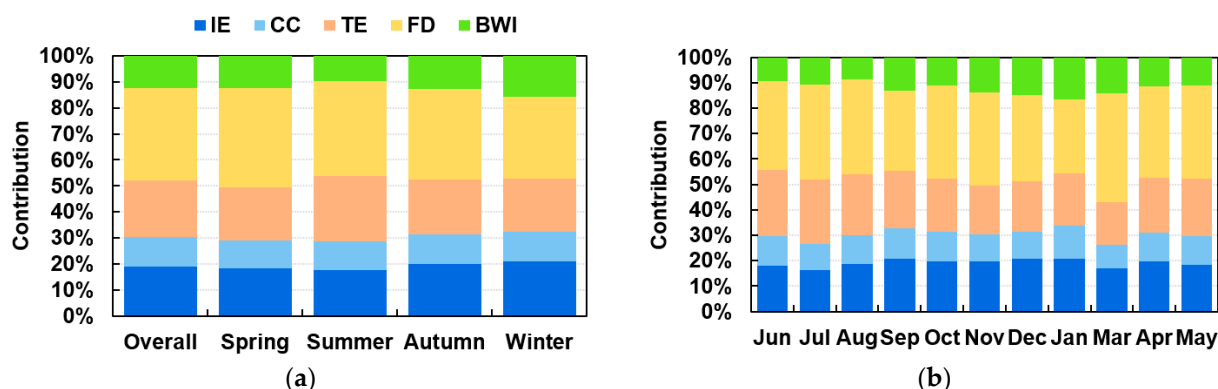


Figure 5. (a) Overall and seasonal variations, and (b) monthly variations in source contributions to trace elements identified via the PMF model.

A previous study conducted in 2014–2015 reported that the primary sources of $PM_{2.5}$ -bound trace elements in Wuhan were CC and IE, with their combined contribution exceeding 40%, while the contribution of FD was merely 9.2% [25]. Despite variations in sampling designs and analytical methods across studies, the reduction in trace element concentrations and the discernible shift in source contributions observed in this study suggest that Wuhan's air pollution control measures over the past decade have actively mitigated anthropogenic emissions amidst rapid urban development. On the other hand, the elevated FD contribution in Wuhan and other cities highlights that fugitive dust pollution remains a formidable challenge for ongoing air quality improvement in China [38,66].

During the sampling period, five sources of trace elements exhibited distinct seasonal trends (Figure 5b). The contribution of FD peaked in spring (38.4%), which coincides with the seasonal maximum in crustal element concentrations (Figure 2). Based on backward trajectory cluster analysis (Figure 6), airflow trajectories passing through arid regions of Mongolia and China during the sampling period in March accounted for a significantly higher proportion (33%) compared to April (5%) and May (9%). The contribution of FD correspondingly peaked in March (43.1%; Figure 5b), corroborating the potential role of long-range dust transport in exacerbating fugitive dust pollution in Wuhan.

The IE and BWI contributions (Figure 5a) exhibited seasonal patterns consistent with those of heavy metal concentrations (Figure 2), with the highest in winter (IE: 20.8%; BWI: 15.8%) and the lowest in summer (IE: 17.6%; BWI: 9.7%). CC maintained relatively stable contributions across seasons (10.6–11.7%), while its peak also occurred in winter (Figure 5a). During the sampling periods in December and January, airflow trajectories originating from Western Siberia, Mongolia, and northern China accounted for 66% and 51% of the total, respectively (Figure 6). Coal-burning hotspots and industrial areas are distributed across Mongolia, Inner Mongolia, Shaanxi, Shanxi, Shandong, and Hebei, where $PM_{2.5}$ is rich in Zn, Pb, As, and Mn [15,18,67]. Air masses passing through these areas likely transport pollutants to Wuhan, thereby increasing the contributions of IE and CC and heavy metal concentrations in winter.

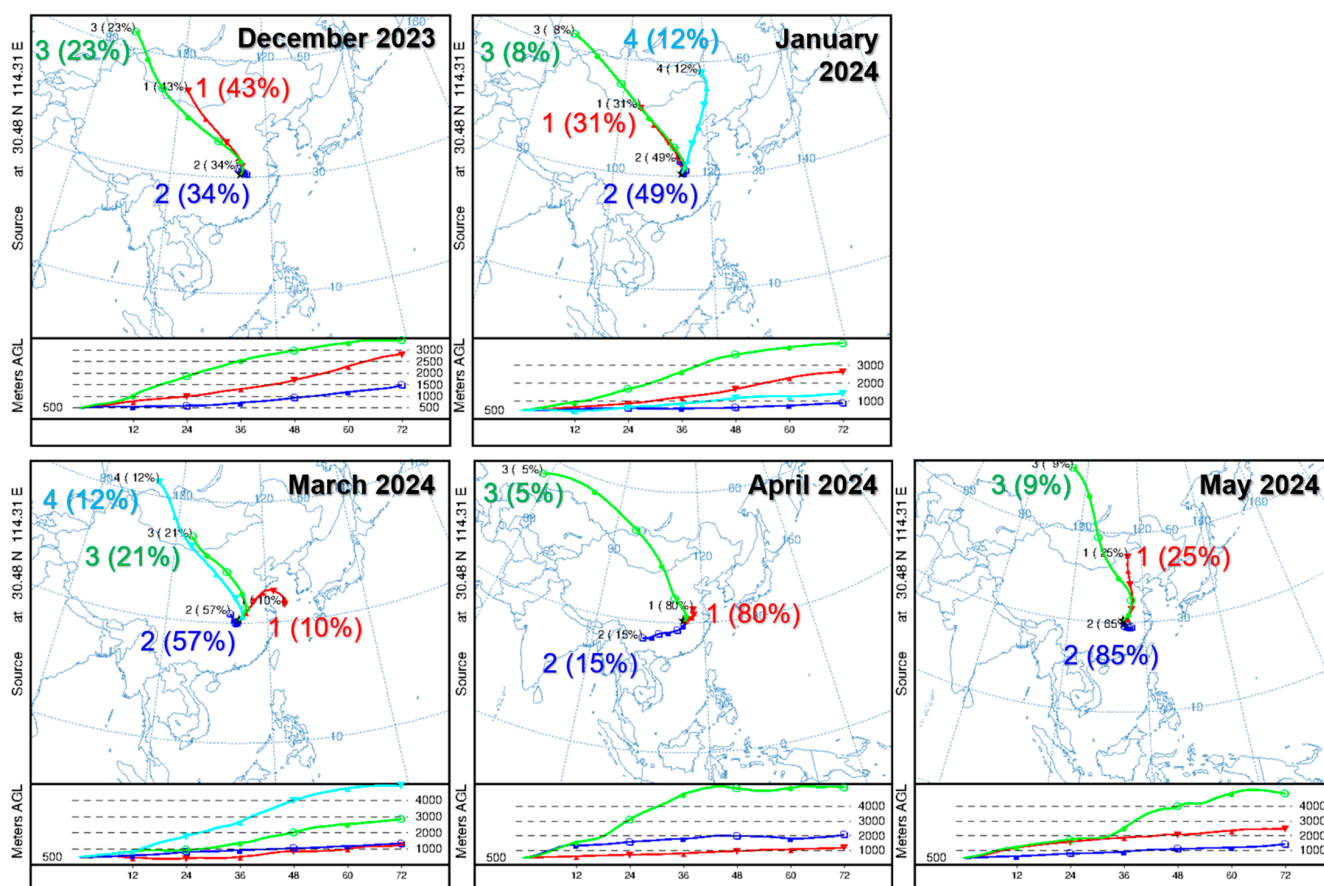


Figure 6. Cluster analysis of 72 h backward trajectories arriving at the sampling site from December to May during the sampling period.

As the remote sensing data from the sampling period of this study were not publicly available, data from 2019 were used as a reference [68]. Nevertheless, the agricultural harvest cycle results in a stable seasonal distribution of biomass burning in central China [69], consistent with the elevated K concentrations observed during autumn and winter in this study. Therefore, the 2019 remote sensing data can reasonably support the analysis of biomass burning sources. Remote sensing monitoring revealed that biomass burning activities in Hubei Province in 2019 were mainly distributed in Wuhan and surrounding cities, with 732 fire hotspots in Wuhan, 1164 in Xiaogan, 530 in Huangshi, and 468 in Huanggang [68,69]. Notably, the monthly variation in biomass burning fire hotspots (Figure S6) presented consistent trends with the contribution of BWI (Figure 5b). Both parameters peaked in January, corresponding to 978 fire hotspots and a contribution of 16.8%. Relatively high values were also observed in March, September, and December, with hotspots ranging from 558 to 716 and contributions ranging from 13.1% to 14.8%. These findings suggest that active biomass burning activities in Wuhan and surrounding cities were primarily responsible for the elevated BWI contribution in winter. Furthermore, abating biomass burning activities (Figure S6) and favorable atmospheric diffusion conditions in summer (Table S6) collectively reduced the contributions of IE, CC, and BWI compared to autumn and winter. Conversely, the contribution of TE increased in summer (Figure 5a). The characteristic elemental ratios, Zn/Pb and Cu/Cd, also reached their annual peaks in summer (4.6 and 0.65, respectively), further corroborating strengthened emissions from gasoline and diesel combustion [3,70]. This trend potentially arises from amplified direct impacts of nearby traffic emissions on the sampling site under improved air quality conditions in summer.

3.4. Health Risks Related to Trace Elements

Figure 7 presents the non-carcinogenic and carcinogenic risks posed to the local population by PM_{2.5}-bound trace elements via inhalation exposure. The *HQ* values of each element for both children and adults ranged from 0.0002 to 0.58 (Figure 7a), below the non-carcinogenic risk threshold (*HQ* = 1) [32]. However, the *HI* value (1.28) of 9 trace elements exceeded 1, with Mn (0.58) and As (0.41) contributing comparatively high *HQ* values, indicating that long-term inhalation exposure may trigger neurological and cardiovascular damage [5,6,9]. Therefore, relevant authorities should prioritize strengthened pollution control and risk mitigation strategies. In addition, due to the algebraic cancellation of exposure terms in risk estimation, identical *HQ* values were obtained for adults and children [71]. Nevertheless, children generally exhibit distinct activity patterns, exposure behaviors, and physiological sensitivity compared with adults. Future research should adopt age-stratified exposure parameters based on field investigations to advance refined health risk assessments for specific populations.

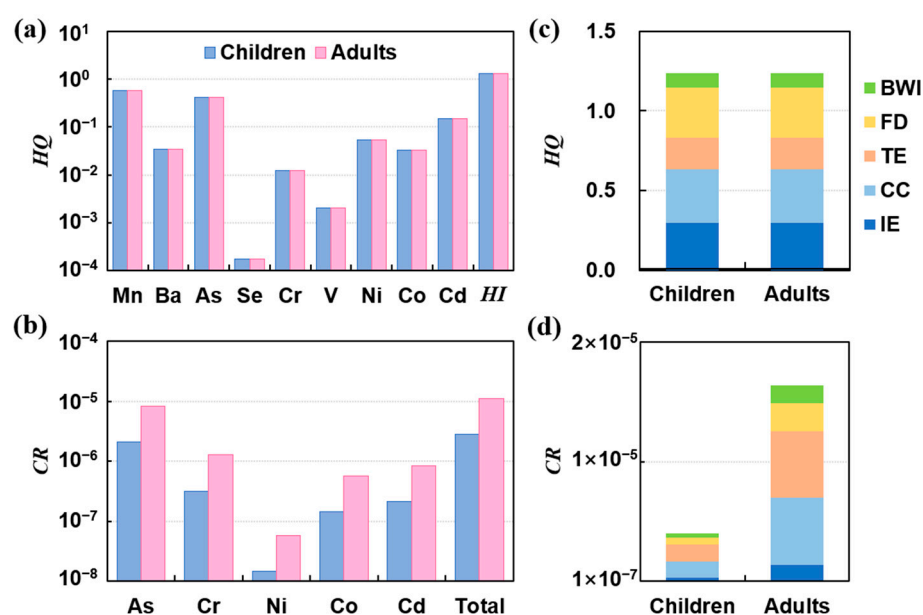


Figure 7. Non-carcinogenic risks (*HQ*) and carcinogenic risks (*CR*) of (a,b) trace elements and (c,d) main sources to local children and adults. *HI* represents cumulative *HQ*.

The cumulative *CR* from trace element exposure was 2.78×10^{-6} for children and 1.11×10^{-5} for adults (Figure 7b). These values are lower than those reported in recent studies in other cities, such as Zhejiang (1.71×10^{-4}), Changzhou (5.87×10^{-5}), Zhengzhou (2.5×10^{-5}), and Baoji (children: 6.81×10^{-6} ; adults: 2.72×10^{-5}) [11,37,42,71], but still exceed the acceptable risk level ($CR = 10^{-6}$) [31]. The *CR* to children was primarily driven by As (2.09×10^{-6}), while that to adults originated from As (8.37×10^{-6}) and Cr (1.27×10^{-6}). Previous studies have also identified As and Cr as major risk factors for elevated *CR*, underscoring the imperative for strengthened management to control their emissions [11,37].

The contributions of the five sources to *HI* and *CR* were estimated based on the PMF model outputs. As shown in Figure 7c, CC, FD, and IE represented the primary contributors to non-carcinogenic risks for both children and adults, accounting for 27.4%, 25.3%, and 23.9% of the *HI* values, respectively. In comparison, VE (16.0%) and BWI (7.5%) exhibited lower contributions. Regarding the *CR* for children and adults (Figure 7d), CC (34.0%) and VE (33.8%) contributed the most, followed by FD (14.1%), BWI (9.1%), and IE (9.0%). Notably, the *CR* for children from CC (1.40×10^{-6}) and VE (1.39×10^{-6}) exceeded

the acceptable risk level, while the CR for adults from all five sources (1.48×10^{-6} to 5.60×10^{-6}) remained above 10^{-6} [31]. These results indicate that pollution reduction measures should be implemented in Wuhan to mitigate trace metal emissions and the associated health risks.

Although CC contributed less to total trace metal concentrations than other sources (Figure 5a), it was the largest contributor to both non-carcinogenic and carcinogenic risks (Figure 7c,d). Coal is primarily used as fuel in coal-fired power plants and industrial enterprises in Wuhan [25]. In recent years, Wuhan has implemented measures such as phasing out coal-fired boilers, promoting clean energy transformation, and enhancing pollution abatement to address CC emissions, and has achieved phased results [21]. Nevertheless, from a health benefits perspective, this study suggests that Wuhan should prioritize coal combustion as a pivotal emission source in future air quality management strategies.

4. Conclusions

This study systematically analyzed the temporal variations, primary sources, and associated health risks of PM_{2.5}-bound trace elements in Wuhan. During the study period, the concentrations of As and Cr exceeded corresponding standards, indicating persistent health risks from toxic trace elements in PM_{2.5} in Wuhan. Local weather conditions, anthropogenic emissions, and regional transport of dust and pollutants caused seasonal variations in trace element concentrations, with crustal elements and heavy metals peaking in spring and winter, respectively. Owing to increased traffic activity and anthropogenic disturbances, trace element concentrations were more abundant on weekdays than weekends, and were mostly higher during daytime than nighttime. Comprehensive air pollution control measures in Wuhan over the past decade have reduced the contribution of the main anthropogenic sources (TE, IE, BWI, and CC) to trace elements, reflecting progress in curbing anthropogenic emissions amid rapid urbanization. Nevertheless, the substantial contribution of FD has made it the primary source of trace elements across all seasons, underscoring the challenge of dust pollution to current air quality improvement. Furthermore, the cumulative non-carcinogenic and carcinogenic risks of trace elements in PM_{2.5} via inhalation exposure exceeded acceptable levels for both local children and adults. Although CC contributed less to trace element concentrations than other sources, it was the largest contributor to non-carcinogenic and carcinogenic risks, followed by FD (to non-carcinogenic risk) and TE (to carcinogenic risk). These findings emphasize the necessity of formulating health-oriented and source-specific policies for air quality management and public health protection. Wuhan should prioritize controlling coal combustion emissions while continuously strengthening traffic regulation and fugitive dust suppression to mitigate trace metal pollution and the associated health risks.

Supplementary Materials: The following supporting information can be downloaded at <https://www.mdpi.com/article/10.3390/atmos17080726/s1>, Text S1: Quality assurance and quality control; Text S2: Trace elements analyses; Text S3: Positive matrix factorization modeling; Table S1: Specific dates for PM_{2.5} sampling in this study; Table S2: Method determination limit (MDL) and linear range for calibration curve of individual trace element, and the chronic inhalation reference concentration (RfC) and inhalation unit risk (IUR) for related trace elements; Table S3: The Q/Q_{exp} ratios of PMF modeling across 3-factor to 7-factor configurations; Table S4: Error estimation results of the best-provided base run in the 5-factor solution; Table S5: Mean concentrations of PM_{2.5}-bound trace elements in Chinese cities and regions reported in previous studies; Table S6: Overall and seasonal mean concentrations and standard deviations of weather parameters including temperature (T), relative humidity (RH), wind speed (WS), rainfall (RF), pressure (P), and boundary layer height (BLH), in Wuhan during the sampling period; Figure S1: Changes in the concentrations of (a) crustal elements and heavy metals and (b) individual crustal elements from March 18 to 20, 2024; Figure S2:

Mean daytime and nighttime concentrations of each trace element in different seasons during the sampling period; Figure S3: Wind rose diagrams of daytime and nighttime concentrations of (a) K, (b) Cu, (c) Bi, and (d) Ba during the sampling period. Grey dashed concentric circles represent different wind speed magnitudes. Solid colored dots denote elemental concentrations corresponding to each wind direction, and the color bar illustrates the gradient of elemental concentration values; Figure S4: Factor profiles of PMF modeling output; Figure S5: Wind rose diagrams of absolute contributions of (a) factor 1, (b) factor 2, (c) factor 3, and (d) factor 4 during the sampling period. Grey dashed concentric circles represent different wind speed magnitudes. Solid colored dots denote factor contributions corresponding to each wind direction, and the color bar illustrates the gradient of factor contribution values; Figure S6: Monthly variations in biomass burning fire hotspots within Hubei province based on remote sensing observations. Data cited from [19]. References [3,11,13,15,19,29,32,35–43,68,71,72] are cited in the Supplementary Materials.

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