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ORIGINAL PAPER

Pollution characteristics and health risk assessment of heavy metals in PM2.5 in Fuxin, China

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Abstract Fuxin is located in the atmospheric channel around Bohai Bay, and its geographical location is very special. Few existing studies have investigated the pollution characteristics and health risk assessment of heavy metals in atmospheric $PM_{2.5}$ during the four seasons in Fuxin, so a total of 180 $PM_{2.5}$ samples were collected from four sampling sites in Fuxin from December 2021 to November 2022. The seasonal distribution characteristics of V, Cr, Mn, Co, Ni, Cu, Zn, Pb, As, Sb, Cd and Ba were analysed via inductively coupled plasma-mass spectrometry (ICP−MS), and the source of the heavy metals was analysed via the enrichment factor (EF) and principal component analysis (PCA). A health risk model was used to assess the health risk of respiratory exposure in men, women and children in Fuxin. The results revealed that the annual average mass order of heavy metals in Fuxin $PM_{2.5}$ was Zn (0.2947 $\mu g \cdot m^{-3}$) > Pb (0.0664 $\mu g \cdot m^{-3}$) > As (0.0225 $\mu g \cdot m^{-3}$) > Ba (0.0205 $\mu g \cdot m^{-3}$) > Mn (0.0187 $\mu g \cdot m^{-3}$) > Cu (0.0140 $\mu g \cdot m^{-3}$) > Cr (0.0095 $\mu g \cdot m^{-3}$) > V (0.0067 $\mu g \cdot m^{-3}$) > Ni (0.0061 $\mu g \cdot m^{-3}$) > Sb (0.0024 $\mu g \cdot m^{-3}$) > Cd (0.0019 $\mu g \cdot m^{-3}$) > Co (0.0007 $\mu g \cdot m^{-3}$). The annual average concentration of As was 3.75 times the GB3095-2012 (China) secondary standard limit, and the concentration of hazard quotient (HQ) in $PM_{2.5}$ was lower than 1, but the concentration of incremental lifetime cancer risk (ILCR) in As was higher than the cancer risk threshold (10^{-4}). These findings indicate a certain risk of cancer in the urban population of Fuxin. Therefore, it is necessary to control the emissions created from coal burning to minimize the health risks to the people of Fuxin.

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Introduction

With the continuous development of cities, the air quality problems caused by industrialisation are becoming more and more serious. Among them, atmospheric $PM_{2.5}$ has triggered the attention of many scholars. Air pollution has gained much attention because it has severe long-term effects on the environment as well as on public health. The factors that influence air pollution include NO_x, CO_x, SO_2, ozone and particulate matter. In particular, the consequences of particulate matter are expanding since particulate matter is stimulated to be adsorbed and ultimately settles on the respiratory or circulatory system of humans. Particulate matter with a diameter of no more than 2.5 µm ($PM_{2.5}$) is the major source of air particulate pollutants and has attracted much attention in recent years. (Balram et al., 2022; Guan et al., 2018; Min et al., 2024; Talbi et al., 2017). According to a report from the WHO, 3.7 million premature deaths annually are associated with outdoor pollution, especially related to $PM_{2.5}$. $PM_{2.5}$ not only causes environmental problems such as smog (Cheng et al., 2016; He et al., 2013; Kumar et al., 2023; Sawaeng et al., 2024) but can also enter the human body because of its small particle size and thus has a direct impact on human health. Some studies have shown that $PM_{2.5}$ present in the atmosphere can enter the human body through blood circulation and have a direct impact on the human respiratory and nervous systems, which results in increased human morbidity (Kioumortzoglou et al. 2016; Samet rt al. 2000; Raaschou et al., 2013; HEI, 2004; Hao et al., 2024; Lal et al., 2023). $PM_{2.5}$ present in the atmosphere comes mainly from anthropogenic sources such as transportation, industrial emissions, and fuel combustion (Alhelí et al., 2024; Sanguineti et al., 2020).

Many researchers have shown that the effects of $PM_{2.5}$ on the human body are related not only to its own concentration but also to some kinds of heavy metals present in the atmosphere (Hao et al., 2018; Pan et al., 2023; Wang et al., 2019). Heavy metals in $PM_{2.5}$ accumulate in the human body and cause various types of diseases after they enter the human body (Sun et al., 2015; Wang et al., 2016); therefore, heavy metals in $PM_{2.5}$ are also the main objects studied by many researchers at home and abroad. Current studies show that heavy metals in $PM_{2.5}$ are derived mainly from industrial sources and road moving sources (Alolayan et al., 2013; Kermani et al., 2018; Maina et al., 2018; Massey et al., 2013).

As a typical coal resource city in northern China, Fuxin is located in the western low mountain and hilly area of Liaoning Province, bordering the Horqin Left Back Banner Sandy Land of the Inner Mongolia Plateau and the Liaohe Plain of Northeast China. The meteorological dynamic conditions are extremely unstable and serve as important atmospheric links east of the Hu Huanyong line between the Horqin Sandy Land in northern China and Bohai Bay in southern China (Wang et al., 2023; Zhang & Pan, 2020). The seasonal absence of farmland soil is obvious, and the synergistic effect of coal-burning soot and northern Horqin aeolian dust during the heating season strongly affects the quality of the atmospheric environment.The climate of Fuxin City belongs to the semi-arid continental monsoon climate of the northern temperate zone, with an average annual temperature of 8.9 °C, and is mainly characterised by excessive precipitation, but with an extremely uneven spatial and temporal distribution, with an average annual precipitation of 591.0 mm (487.3 mm in a calendar year), and the longest continuous precipitation in a year of 4 d. The air temperature is on the high side, and there is plenty of sunshine, with the highest annual air temperature reaching 35.9 °C. The Haizhou opencast coal mine in Fuxin is famous worldwide. From 1953 to 2005, large-scale mining stopped production, and half a century of mining formed an enormous opencast dumping pit with a volume of approximately 4 billion m^3 and a waste dump with a volume of nearly 850 million m^3. At the same time, it is only 3 km south of the urban area. Many studies have shown that mines contribute significantly to atmospheric dust and heavy metal pollution in urban areas (Patel et al., 2012; Zhao et al., 2017a, 2017b; Zhou et al., 2023). There are very few reports on the characteristics of $PM_{2.5}$ and heavy metal pollution in Fuxin. The 2022 Winter Olympics, held in Beijing, will last for 17 days, starting on 4 February and ending on 20 February. Fuxin is in the atmospheric channel and affects Beijing's air quality. Therefore, in order to ensure the air quality in Beijing in the future, it is of high practical significance to study the pollution characteristics and health risk assessment of heavy metals in atmospheric $PM_{2.5}$ in Fuxin City.

Materials and methods

Sample collection

In this study, four $PM_{2.5}$ sampling sites were set up in Fuxin, as shown in Fig. 1, and the routine sites were located at the Fuxin Environmental Monitoring Center (121° 40′37.6″E, 42°01′28.7″N). The temporary monitoring points include a coal quality laboratory (121°40′14.8″ E, 42° 01′12.2″ N), a comprehensive performance monitoring station (121°40′13.2″ E, 42°01′17.9″ N) and a grain and oil monitoring station (121°39′08.6″ E, 41°59′55.9″N). According to the national environmental protection standards of the People's Republic of China (HJ618–2011), the height of the sampling instruments is 10 m.

$PM_{2.5}$ samples were collected from December 2021 to February 2022 (winter), March 2022 to May 2022 (spring), June 2022 to August 2022 (summer), and September 2022 to November 2022 (autumn) with a medium-flow particulate matter sampler (Model lao1108a-1). This sampler is widely used in atmospheric sampling. The sampler was 48 cm long, 40 cm wide and 1 m high. The sampling time was set from 9:00 AM on the same day to 8:30 AM on the next day, and the time was 23.5 h. The flow rate was 100 L min^{-1}. A total of 180 valid samples were collected. The main meteorological parameters measured during the sampling period are shown in Table 1.

Sample analysis

In accordance with the "technical specification for manual monitoring method (gravimetric method) of ambient air particulate matter ($PM_{2.5}$), the filter membrane was balanced in the environment at a temperature of 20 ± 1 °C and humidity of 50% ± 1% for 48 h before and after sampling. A 1/4 polypropylene filter membrane was cut with ceramic scissors and placed in the digestion tank, after which 5 ml of nitric acid (pH = 5.6) and 0.05 ml of 40% HF (pH = 5.3) were added. After these acids were added, they were

Table 1 Average temperature, humidity and wind speed during sampling

Seasons	Temperature/°C	Humidity/%	Wind Speed/(m/s)
winter	-9.27	38.13	1.38
spring	0.93	36.8	1.74
summer	26.09	40.2	3.18
autumn	18.6	64.8	1.8

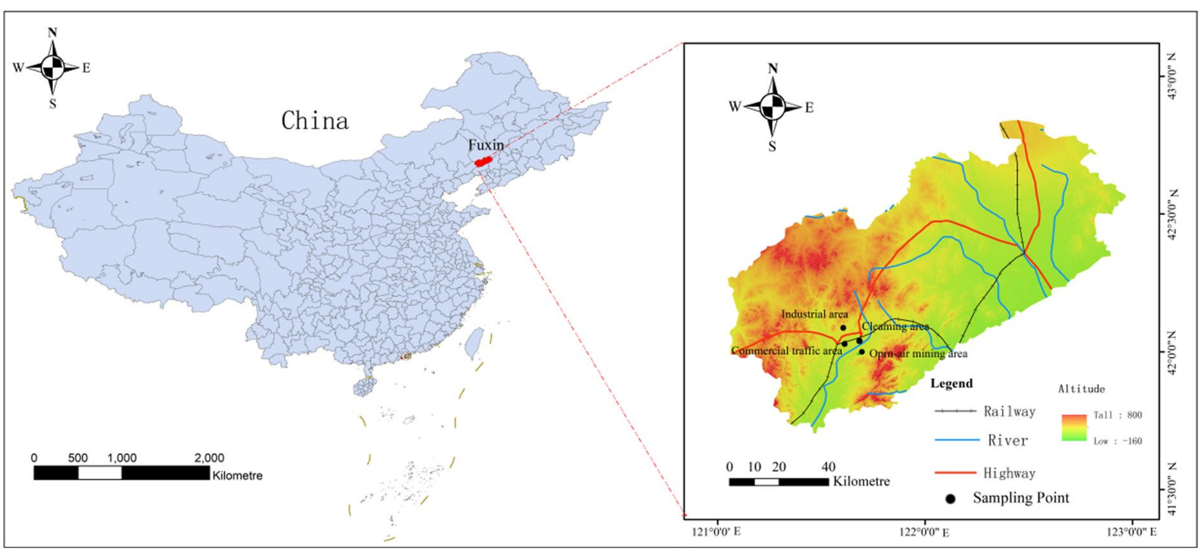

Fig. 1 Distribution of sampling points in Fuxin

dissolved properly and refluxed at 220 °C for 2 h. Then, 5 ml of dilute nitric acid (pH = 5.4) was added, and the solution was transferred to 10 ml. Twelve metals, such as V, Cr, Mn, Co, Ni, Cu, Zn, Pb, As, Sb, Cd and Ba (Zhao et al., 2020a, 2020b), were analysed via ICP−MS.At least one standard sample must be analysed for each group of samples, operated and calculated at recoveries of 80% to 120%; at least one duplicate sample must be analysed for each group of samples, with a relative error of less than 20%; a blank sample must be prepared for each group, and the value of the test must be less than or equal to twice the limit of detection.

The membrane was changed before and after each sampling to ensure that the filter membrane was flat and free of burrs and damage. The sampling head was cleaned once every 168 h. For every 10 samples measured, a blank filter membrane was used. Single-point calibration was conducted to ensure that the blank control samples and quality control samples in each batch of experiments were measured synchronously. Each batch (≤ 20) was tested for spiked recovery. The recovery, average relative standard deviation (RSD) and standard curve R^2 of the 8 water-soluble ions were 95.5 ~ 105.5%, < 10% and 0.999, respectively.

Results and discussion

Temporal distribution characteristics of the $PM_{2.5}$ concentration

Measurements of $PM_{2.5}$ in the atmosphere were taken synchronously at four sampling sites in Fuxin via the gravimetric method. The results are shown in Fig. 2. The average annual concentration of $PM_{2.5}$ was 39.68 μg·m^{-3} in Fuxin during the sampling period from December 2021 to November 2022. Among them, 176 days exceeded the safe concentration limit (10 μg·m^{-3}) of $PM_{2.5}$ prescribed by the World Health Organization, accounting for 97.8% of the total sampling period. Approximately 80% of the days exceeded the daily average concentration limit of $PM_{2.5}$ (35 μg·m^{-3}) set by the United States, and 16 days exceeded the daily average concentration standard of $PM_{2.5}$ (75 μg·m^{-3}) set by China, accounting for 8.9% of the total sampling days.

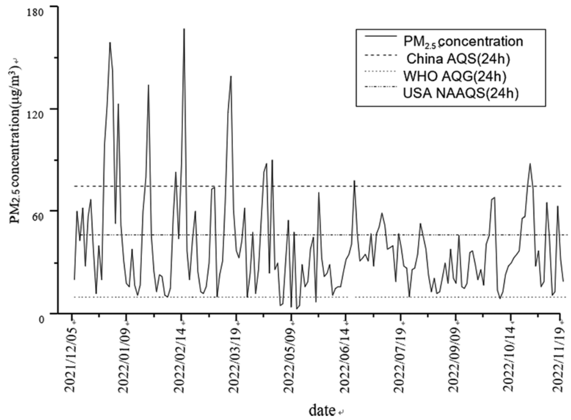

Fig. 2 Daily average $PM_{2.5}$ concentration in Fuxin during sampling period

These findings indicate that $PM_{2.5}$ pollution in Fuxin has improved (Zhao et al., 2015). The average mass concentration of $PM_{2.5}$ was 52.93 μg·m^{-3} in winter and 39.18 μg·m^{-3} in spring. The average mass concentration of $PM_{2.5}$ in spring was approximately 26% lower than that in winter because heating stopped in Fuxin at the end of March. The average mass concentrations of $PM_{2.5}$ in summer and autumn were 32.29 μg·m^{-3} and 34.31 μg·m^{-3}, respectively. The results were the same as those in Lanzhou and Xi'an (Li et al., 2018; Liu et al., 2023). The highest concentration of $PM_{2.5}$ in Fuxin was recorded on December 18 (159 μg·m^{-3}), with an average daily humidity of 96%, an air quality index (AQI) of 209 and a prevailing wind direction of 1.02 m/s. High humidity and low wind speed provide meteorological conditions for high $PM_{2.5}$ pollution. Owing to the special geographical location in Fuxin, the wind direction has a great influence on the concentration of $PM_{2.5}$. During the sampling period, the lowest $PM_{2.5}$ concentration in Fuxin was found on May 11, with no rainfall and a relative humidity of 44%, and the $PM_{2.5}$ concentration decreased to 3 μg·m^{-3}, which was mainly due to the effects of "rain in the cloud" on $PM_{2.5}$ "nuclear condensation" and "scoured below the cloud" on $PM_{2.5}$ "collisional coagulation" (Li et al., 2014; Xuan, Xue, and Lei 2019; Han et al., 2017;Bui et al., 2023).

Influencing factors of the $PM_{2.5}$ concentration

Figure 3 clearly shows that the annual variation trend of the $PM_{2.5}$ concentration with NO_2 and SO_2 was stable and relatively synchronous, and the correlation coefficients of $PM_{2.5}$ with NO_2 and SO_2 were 0.777 and 0.655, respectively, during the sampling period. The industrial structure of Fuxin is relatively singular, and the population in the urban area is only 600,000. Coal-fired smoke emissions from power plants and thermal power plants constitute the main source of $PM_{2.5}$. NO_2 and SO_2 in the urban area of Fuxin and the concentrations of these three pollutants are relatively high in winter and spring and relatively low in summer and autumn. There is a large concentration gradient with the same trend between the three pollutant concentrations in summer and autumn, which is due mainly to the lack of important emission sources for heating coal combustion. The emissions of $PM_{2.5}$ and SO_2 are greatly reduced, and the emission of NO_2 is due to the contribution of motor vehicle exhaust; compared with those in the winter and spring seasons, NO_2 concentrations do not decrease significantly. Moreover, gaseous precursors such as SO_2 and NO_2 can produce secondary pollutants such as sulfate and nitrate aerosols through homogeneous or heterogeneous (particle surface) reactions, which can increase the concentration of $PM_{2.5}$(Ambade et al., 2020; Cao et al., 2024; Zhe et al., 2022).

The variation in the $PM_{2.5}$ concentration in winter and spring in Fuxin was greater than that in summer and autumn, and the variation in the O_3 concentration in winter and spring was smaller than that in winter and spring. The correlation coefficients between the $PM_{2.5}$ concentration, temperature and O_3 concentration were -0.166 and -0.038, respectively. In summer and autumn, the surface temperature is higher than the atmospheric temperature, and atmospheric convection is favourable for the diffusion and dilution of $PM_{2.5}$.

The results revealed that evergreen leaves could effectively retain $PM_{2.5}$ (Yang et al., 2018; Zeng et al., 2023). Moreover, the amount of light radiation increases because of the lower concentration of $PM_{2.5}$. Therefore, it can easily excite the photochemical chain reaction of NO_2 and other tail gases of motor vehicles and increase the concentration of O_3. Previous studies have shown that the concentrations of $PM_{2.5}$ and O_3 in Fuxin have a staggered peak relationship (Zhao et al., 2021).

The northern part of Fuxin is the Horqin Sandy Land, bordered by the Liaohe Plain to the east, Nuerhu Mountain to the west, and the southern part is linked to Bohai Bay. It is a transitional zone between the Inner Mongolian steppe and the Rocky Mountains of North China. It presents a semienclosed hilly basin landform. The wind roses during sampling are shown in Fig. 4. Southwesterly winds are the dominant wind

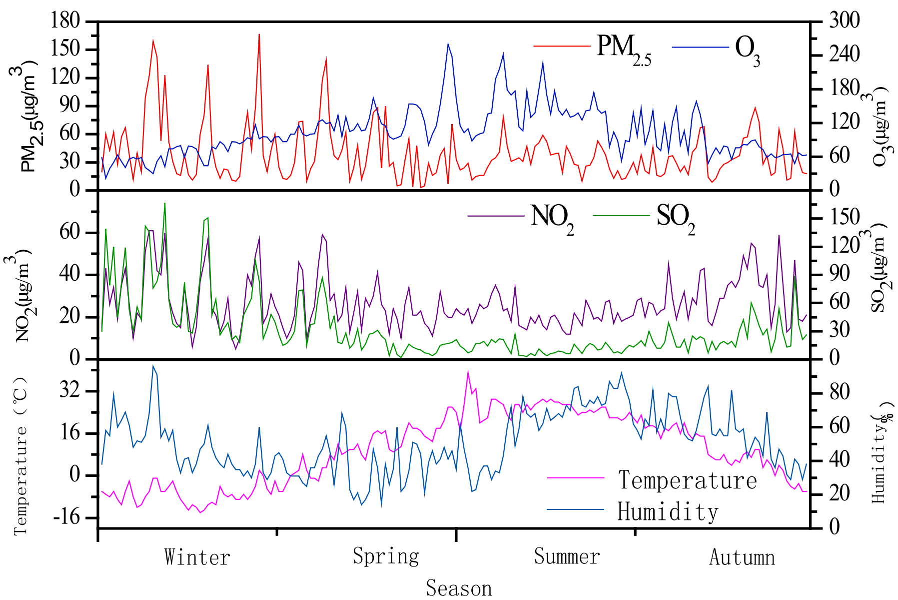

Fig. 3 Change of meteorological conditions during sampling period

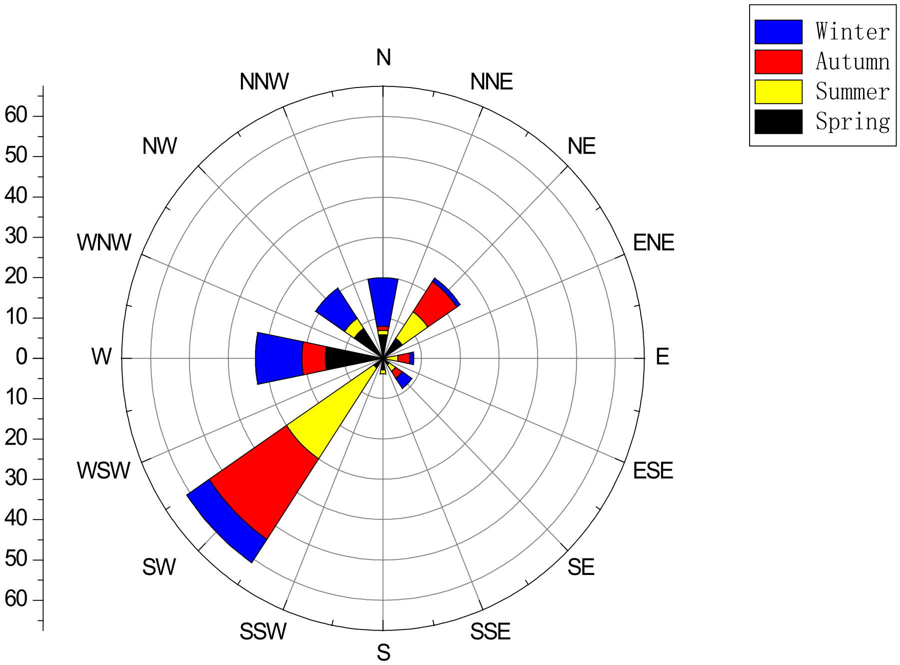

Fig. 4 Wind rose map of fuxin city

direction in Fuxin and significantly contribute to the rapid accumulation of $PM_{2.5}$ in Fuxin (Zhao et al., 2020a, 2020b).

According to the wind roses of Fuxin during the sampling period, the prevailing winds in the Fuxin region in winter and spring were northerly and westerly, and the particulate matter in the Horqin Sandy Land, the largest sandy land in the northwestern world, contributed directly to the $PM_{2.5}$ concentration in the Fuxin region. The phenomenon of temperature inversion, which is not conducive to the diffusion of pollutants and can promote an increase in the atmospheric $PM_{2.5}$ concentration, is easy. The wind direction directly affects the air humidity and temperature in Fuxin, affects the secondary conversion of gaseous precursors such as SO_2 and NO_2, and affects the hygroscopic growth of $PM_{2.5}$ particles and fluctuations in the $PM_{2.5}$ concentration.

Concentration levels and time distributions of heavy metals in $PM_{2.5}$

The measured results of the mass concentrations of heavy metals in atmospheric $PM_{2.5}$ in Fuxin city during the sampling period are shown in Fig. 5, and their average annual concentrations from high to low were as follows: Zn (0.2947 μg·m^{-3}) > Pb

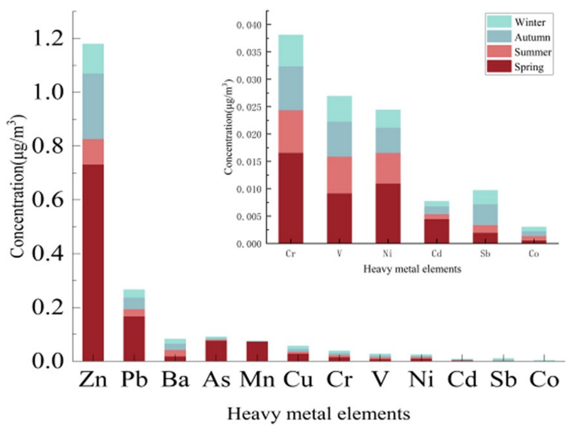

Fig. 5 Measurement of heavy metals concentration in $PM_{2.5}$ of fuxin city

(0.0664 μg·m^{-3}) > As (0.0225 μg·m^{-3}) > Ba (0.0205 μg·m^{-3}) > Mn (0.0187 μg·m^{-3}) > Cu (0.0140 μg·m^{-3}) > Cr (0.0095 μg·m^{-3}) > V (0.0067 μg·m^{-3}) > Ni (0.0061 μg·m^{-3}) > Sb (0.0024 μg·m^{-3}) > Cd (0.0019 μg·m^{-3}) > Co (0.0007 μg·m^{-3}). The average concentrations of Pb and As are 1.2 and 3.75 times the GB3095-2012 secondary standard limits, respectively, which are 3 and 1.13 times the EU air standard limits. Pb and As can enter the human body, resulting in several diseases related to the respiratory system. Compared with those in other cities in Fushun, Jinzhou, Panjin and Anshan (Gu et al., 2016; Li, 2017; Li et al., 2019; Wang et al., 2017), the concentrations of Cr in $PM_{2.5}$ in Fuxin are more than 1.69, 2.31, 2.25 and 1.13 times greater than those in Fushun, Jinzhou, Panjin and Anshan, respectively. The possible reason for the increased concentration of Cr is the existence of the leather industry, which is responsible for economic growth in Fuxin. Many chrome alum and dichromate materials are used in the leather industry, which discharge many leather tanning materials. In Fuxin, this leather industry is located in Xinqiu District, which is 6 km from the city (Xie, Hou, and Chen 2018).

The variation ranges of Zn, Pb, As and Mn with respect to the seasonal concentration were relatively high, and the maximum average concentration occurred in spring, whereas the minimum concentration occurred in summer. With the exception of mica, which is the main source of Mn, both Zn and As are associated with industrial processes (Jiao et al., 2014; Tian et al., 2010), with the highest values occurring in spring because of emissions from the coal-fired heating and metal smelting industries. Pb may be closely related to the combustion of fossil fuels in automobiles (Wang et al., 2015), the dominant wind direction in spring is northwest, and Mn may be contributed by the Horqin Sand transported by the northern Horqin Sand Land and southern Bohai Bay Air Passage.

Source apportionment of heavy metals in $PM_{2.5}$

The enrichment factor (EF) method was proposed by Gordon in the 1970s to judge the impact of man-made pollution sources other than natural sources on atmospheric particulate matter. Sakan et al. studied pollution-related heavy metals in freshwater sediment in Serbia, and Kumar et al. used the enrichment factor method to evaluate the extent to which anthropogenic activities affect the concentration of heavy metals in the mangrove forests of south-west India and the sediment of neighbouring estuarine stations(Sakan et al., 2015).Loska K and others believe that although the method has some shortcomings, it has a standardized formula, so it can still be used as a simple and good method to estimate the enrichment level of elements. The formula is as follows:

$$EF = \frac{(C_i/C_n)_{\text{particulate matter}}}{\left(C_i/C_n\right)_{soil}} \tag{1}$$

where C_i is the concentration of the ith element; C_n is the concentration of the selected reference element; the numerator part of the formula represents the amount of elements in particulate matter; and the denominator part represents the amount of elements in the soil. The reference elements are all particulate matter, and the content in the soil is relatively high. The frequently used reference elements are Al, Fe, and Ti. Fe has relatively stable chemical properties and is a commonly used reference element. Therefore, Fe is selected as the reference element in this paper.

Figure 6 shows that the concentration indices of Cd and Zn in the $PM_{2.5}$ of the Fuxin atmosphere were greater than 100 during the sampling period, which indicates that the concentration indices of Cu, As, Sb and Pb were all between 10 and 100. The enrichment indices of V, Cr, and Ni range from 2 ~ 10, which is considered moderate enrichment, which means that there is less man-made influence. However, the enrichment indices of Mn, Co and Ba are less than 2, indicating a slight man-made influence.

The concentration index of heavy metals in $PM_{2.5}$ during winter and spring is generally greater than that in summer and autumn, and the concentration index of Cd in spring is the highest, reaching 920.88 in spring, which is much greater than 100; these findings indicate that the concentration of heavy metals in $PM_{2.5}$ during winter and spring is strongly affected by

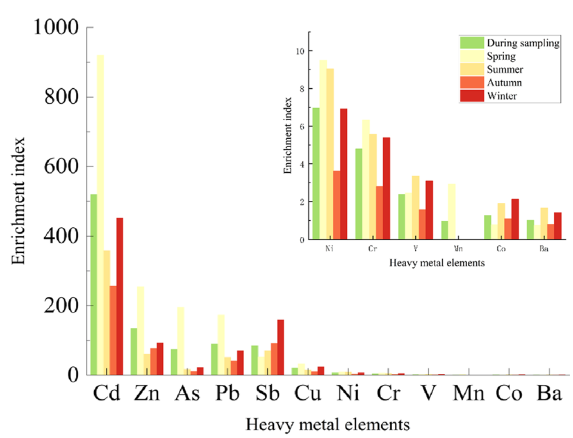

Fig. 6 Concentration index of heavy metals in $PM_{2.5}$ atmosphere of fuxin city

human activities. Zhang Song, You Fang and others have shown that the main source of atmospheric Cd is coal burning (You et al., 2019; Zhang et al., 2020). As a coal resource city with a century-long mining history, Fuxin has five coal-fired thermal power plants, such as Fuxin power generation, Jinshan district coal gangue thermal power, Eagle cement, Jiechao coal gangue thermal power, and Fuxin mining group coal gangue thermal power plants. The enrichment indices of Zn, Pb and As are 254.98, 173.40 and 195.14, respectively, which are very high in spring. The higher enrichment factor in spring is attributed mainly to coal burning and motor vehicle emissions. The number of vehicles in Fuxin continues to grow, currently reaching 300,000, and the annual increase in motor vehicle emissions may be attributed to Zn, Pb and As. Fuxin is prone to the accumulation of air pollutants due to its poor dispersion conditions owing to its "North-facing south" dustpan topography.

In this paper, SPSS22.0 software was used to analyse the source of 12 heavy metal elements in Fuxin City by PCA method, as shown in Tables 2 and 3.

The eigenvalues of the three principal components were 13.826, 1.969 and 1.283 with a cumulative contribution of 95.076 per cent, indicating that the three principal components provided sufficient information for the original data.

The variance contribution of principal factor 1 to atmospheric particulate matter PM2.5 was 77.013%, with high loading values for As and Pb. Stembeck (Sternbeck et al., 2002) found that coal-fired boiler flue gas dust has a high percentage of Pb and As relative to other trace elements, and thus this factor can be considered as a source of coal soot.

The main factor 2 has a variance contribution of 10.937% to atmospheric $PM_{2.5}$ and it has high loading values for Zn and Cd, Begun et al. (Begun et al.2004) found that Zn is mainly derived from tyre and ground abrasion and motor vehicle lubricants, and that the sources of pollution from these two elements are mainly coal combustion and motor vehicle exhaust, so this factor can be considered as a mobile source.

Table 2 Principal component eigenvalues and contributions

Principal component	Eigenvalue	Per cent	Percentage
1	13.826	77.013%	77.013%
2	1.969	10.937%	87.950%
3	1.283	7.126%	95.076%

Table 3 Elemental rotational factor loadings in $PM_{2.5}$

Factor	Principal component		
	1	2	3
Zn	0.760	0.834	0.232
Pb	0.921	0.581	0.114
Ba	-0.014	0.360	0.229
As	0.892	0.480	0.558
Mn	0.461	-0.114	0.708
Cu	-0.088	0.567	0.341
Cr	0.237	0.346	0.403
V	0.425	0.563	0.348
Ni	0.765	0.194	0.569
Cd	0.678	0.795	0.230
Sb	0.439	0.612	-0.119
Co	0.691	0.591	0.348

The variance contribution of main factor 3 to atmospheric $PM_{2.5}$ is 7.126%, with the highest loading value for Mn. From the enrichment factor value, it can be seen that the enrichment value of Mn is not high, and it is subject to low anthropogenic influences, so it can be regarded as the contribution of the Kerqin wind and sand transported by atmospheric channels.

Health risk assessment of heavy metal elements in $PM_{2.5}$

The human health risk assessment model was proposed by the Environmental Protection Agency (EPA) in 1983. The risk assessment is divided into four steps: hazard identification, dose response, exposure assessment and risk characterization (US, 2002). The data collected from the International Cancer Research Institute and the EPA comprehensive risk information show that the pollutants are classified into carcinogens and noncarcinogens.

Based on the US EPA Soil Health Risk Evaluation Model and with reference to the China Population Exposure Parameter Manual, the population health risk of heavy metals in atmospheric precipitation in the urban area of Fuxin City was assessed. Formulae for the calculation of non-carcinogenic risk values and total non-carcinogenic risk for heavy metals in atmospheric dustfall:

$$HQ_{i,j} = ADD_{i,j}/RfD_{i,j} \quad (2)$$

$$HI = \sum_{i=1}^{n} \sum_{i=1}^{m} HQ_{i,j} \quad (3)$$

where: $HQ_{i,j}$ is the Hazard Quotient characterising the health risk of heavy metal i via j; $ADD_{i,j}$ is the daily exposure of heavy metal i via j, mg/(kg·d); $RfD_{i,j}$ is the Hazard Quotient reference dose of heavy metal i via j; $RfD_{i,j}$ is the Hazard Quotient reference dose of heavy metal i via j; $RfD_{i,j}$ is the Hazard Quotient reference dose of heavy metal i via j.$RfD_{i,\ j}$ is the reference dose for Hazard Quotient of heavy metal element i through j route, indicating the maximum amount of heavy metal per unit time and unit body mass that can be ingested by human body without causing adverse reaction, mg /(kg·d);

HI is the sum of Hazard Quotient, when $HQ_{i,\ j}$ or HI is less than 1, it can be regarded as Hazard Quotient of heavy metal in the atmospheric dustfall on human beings is relatively small or can be ignored; when $HQ_{i,\ j}$ or HI is more than 1, then it is regarded as Hazard Quotient of heavy metal in atmospheric dustfall on human beings, and it can be regarded as Hazard Quotient on human beings. When $HQ_{i,\ j}$ or HI is greater than 1, it is considered that heavy metals in atmospheric dustfall pose a Hazard Quotient to human beings and may cause damage to human health.

As the exposure and health risk evaluation process may be affected by the model used for the evaluation, the representativeness of the sample collection, the accuracy of the analysis of the results, and the accuracy of the relevant exposure parameters in the model, there is a certain degree of uncertainty in the results of its health risk evaluation.The 12 heavy metals in $PM_{2.5}$ in the Fuxin atmosphere during the sampling period are shown in Table 4. The heavy metal ions that pose noncarcinogenic risks from high to low risk are Mn, V, Co, Cr, As, Pb, Sb, Cd, Zn, Cu, Ni and Ba. The noncarcinogenic risk coefficient (HQ) of the 12 heavy metal elements was $1.08\times10^{-6}\sim1.85\times10^{-2}$, which was lower than EPA limit 1 (EPA, 1989). The noncarcinogenic risk of heavy metals in Fuxin was generally low during the sampling period, and the risk gradually decreased in males, females and children.

The lifetime cancer risk of the five heavy metals in Fuxin $PM_{2.5}$ ranges from 6.26×10^{-8} to 4.14×10^{-6}. The heavy metals that are associated with high to low cancer risk are As, Cd, Cr, Co, and Ni; among them, As has the highest cancer risk ($4.14\times10^{--6}$), which lies within the range of carcinogenic risk ($10^{-6}\sim10^{-4}$). The results revealed that As in $PM_{2.5}$ posed a carcinogenic risk, and the carcinogenic risk values of the other heavy metal elements were all lower than the threshold of carcinogenic risk. As coal combustion is a symbolic element of coal combustion, it may be related to the relatively single energy structure of coal in Fuxin. The Haizhou Mine, once the largest open pit mine in Asia, is located only 3 km south of the urban area of Fuxin, and there are currently more than 200 sites of spontaneous combustion of residual coal; at the same time, large amounts of coal gangue and fly ash accumulate around the open-pit mine. Under wind disturbance, the concentrations of $PM_{2.5}$

Table 4 Risk of respiratory exposure of heavy metals to $PM_{2.5}$ in fuxin city during sampling period

Heavy metal	HQ Adult male	HQ Adult female	HQ children	SF	ILCR
V	1.33×10^{-2}	1.20×10^{-2}	1.01×10^{-2}		
Cr	4.55×10^{-3}	4.11×10^{-3}	3.46×10^{-3}	0.84	9.76×10^{-8}
Mn	1.85×10^{-2}	1.67×10^{-2}	1.40×10^{-2}		
Co	1.81×10^{-3}	1.64×10^{-3}	1.38×10^{-3}	9.8	8.92×10^{-8}
Ni	4.23×10^{-6}	3.82×10^{-6}	3.21×10^{-6}	0.84	6.26×10^{-8}
Cu	4.83×10^{-6}	4.37×10^{-6}	3.68×10^{-6}		
Zn	1.36×10^{-5}	1.23×10^{-5}	1.03×10^{-5}		
As	1.04×10^{-3}	9.37×10^{-4}	7.88×10^{-4}	15.1	4.14×10^{-6}
Cd	2.66×10^{-5}	2.41×10^{-5}	2.03×10^{-5}	6.3	1.48×10^{-7}
Sb	8.41×10^{-5}	7.60×10^{-5}	6.40×10^{-5}		
Pb	2.62×10^{-4}	2.37×10^{-4}	2.00×10^{-4}		
Ba	1.42×10^{-6}	1.28×10^{-6}	1.08×10^{-6}		

and As, which are the identifying elements of coal-fired sources in urban areas, greatly contribute. The results of the health risk assessment of heavy metal elements in $PM_{2.5}$ in Nanjing and Xi'an revealed that the lifetime cancer risk of As exceeded the threshold range of cancer risk (Zhao, 2016, 2018) and that there is a certain risk of carcinogenesis to the main population in the region. Excessive As can interfere with the normal metabolism of cells, affect the process of respiration and oxidation, cause pathological changes in cells, and eventually cause various diseases.

Therefore, Fuxin should strengthen the total emission control of coal combustion to reduce $PM_{2.5}$ emissions and heavy metals from coal-burning exposure to the health risks of the local population.First of all adjust the industrial structure to implement emission reduction, effectively control the rapid growth of high energy-consuming and high-polluting industries; accelerate the elimination of backward production capacity, focus on the implementation of key governance projects to implement emission reduction; the full implementation of key emission reduction projects, replacing the capacity of small, low-efficiency, heavily polluting industries; grasp the supervision and management of the implementation of emission reduction, strengthen environmental supervision, strengthen the cleaner production audits, and actively guide the orderly development of the circular economy.

Conclusion

(1) During the sampling period, $PM_{2.5}$ in Fuxin City exceeded the daily average secondary concentration standard of $PM_{2.5}$ in China (75 $\mu g \cdot m^{-3}$) and the World Health Organization (WHO) safe concentration limit for $PM_{2.5}$ (10 $\mu g \cdot m^{-3}$) in 8.9% and 97.8% of the total number of sampling days, respectively, indicating that the pollution situation in Fuxin City has not yet reached a safe level and that Fuxin City still needs to control $PM_{2.5}$.

(2) During the sampling period, the concentrations of Pb and As in atmospheric $PM_{2.5}$ in Fuxin exceeded the standards. Compared with that in cities in Liaoning Province, such as Fushun, Jinzhou, Panjin and Anshan, the concentration of Cr in atmospheric $PM_{2.5}$ in Fuxin was significantly greater, which was closely related to the leather industry in Fuxin.

(3) The results of the enrichment index method revealed that the enrichment indices of Cd and Zn in atmospheric $PM_{2.5}$ in Fuxin city were greater than 100 during the sampling period, which was strongly affected by human activities. The enrichment index of heavy metals in atmospheric $PM_{2.5}$ in winter and spring was generally greater than that in summer and autumn, and the sources of heavy metals were mainly combustion sources and motor vehicle emission sources.The results of principal component analysis indicate that the pollution sources are coal soot sources, mobile sources of motor vehicle exhaust, etc., which are mutually verified with the results of the enrichment factor method.

(4) The results of the health risk assessment showed that the risk indices of the five carcinogenic heavy metals were As, Cd, Cr, Co, and Ni, in that order.The health risk value of As exceeded the carcinogenic risk threshold. Therefore, Fuxin City should strengthen the control of total coal emissions to reduce the health risks of local residents from respiratory exposure, ingestion exposure and dermal exposure to heavy metal elements emitted from coal.

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Data availability The datasets generated during and/or analysed during the current study are not publicly available owing to [REASON(S) WHY DATA ARE NOT PUBLIC] but are available from the corresponding author upon reasonable request.].

Declarations

Conflict of interests The authors have no relevant financial or nonfinancial interests to disclose.

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