Chemical characterization, source apportionment, and health risk assessment of PM$_{2.5}$ in a typical industrial region in North China

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Abstract
To clarify the chemical characteristics, source contributions, and health risks of pollution events associated with high PM$_{2.5}$ in typical industrial areas of North China, manual sampling and analysis of PM$_{2.5}$ were conducted in the spring, summer, autumn, and winter of 2019 in Pingyin County, Jinan City, Shandong Province. The results showed that the total concentration of 29 components in PM$_{2.5}$ was $53.4 \pm 43.9$ μg·m$^{-3}$, including OC/EC, water-soluble ions, inorganic elements, and metal elements. The largest contribution was from the NO$_3^-$ ion, at $14.6 \pm 14.2$ μg·m$^{-3}$, followed by organic carbon (OC), SO$_4^{2-}$, and NH$_4^+$, with concentrations of $9.3 \pm 5.5$, $9.1 \pm 6.4$, and $8.1 \pm 6.8$ μg·m$^{-3}$, respectively. The concentrations of OC, NO$_3^-$, and SO$_4^{2-}$ were highest in winter and lowest in summer, whereas the NH$_4^+$ concentration was highest in winter and lowest in spring. Typical heavy metals had higher concentrations in autumn and winter, and lower concentrations in spring and summer. The annual average sulfur oxidation rate (SOR) and nitrogen oxidation rate (NOR) were $0.30 \pm 0.14$ and $0.21 \pm 0.12$, respectively, with the highest SO$_2$ emission and conversion rates in winter, resulting in the SO$_4^{2-}$ concentration being highest in winter. The average concentration of secondary organic carbon in 2019 was $2.8 \pm 1.9$ μg·m$^{-3}$, and it comprised approximately 30% of total OC. The concentrations of 18 elements including Na, Mg, and Al were between $2.3 \pm 1.6$ and $888.1 \pm 415.2$ ng·m$^{-3}$, with Ni having the lowest concentration and K the highest. The health risk assessment for typical heavy metals showed that Pb poses a potential carcinogenic risk for adults, whereas As may pose a carcinogenic risk for adults, children, and adolescents. The non-carcinogenic risk coefficients for all heavy metals were lower than 1.0, indicating that the non-carcinogenic risk was negligible. Positive matrix factorization analysis indicated that coal-burning emissions contributed the largest fraction of PM$_{2.5}$, accounting for 35.9% of the total. The contribution of automotive emissions is similar to that of coal, at 32.1%. The third-largest contributor was industrial sources, which accounted for 17.2%. The contributions of dust and other emissions sources to PM$_{2.5}$ were 8.4% and 6.4%, respectively. This study provides reference data for policymakers to improve the air quality in the NCP.

Keywords North China · PM$_{2.5}$ · Chemical composition · Source apportionment · SOC · Health risk assessment
Introduction

The North China Plain (NCP) is one of the most heavily atmospherically polluted areas in China. From a geographical perspective, the North China Plain is located between the Yanshan Mountains in the north, Taihang Mountains in the west, and Bohai Bay in the east, which create a semi-closed topography with high-elevation terrain in the northwest and low-elevation terrain in the southeast. Due to this configuration, air pollutants are easily transmitted into the area, but do not readily diffuse out. Heavy emissions from industrial processes, coal-burning emissions from energy production, and traffic emissions in North China contribute to the high emission levels of atmospheric pollutants (Jia et al. 2021; Luo et al. 2021; Wu et al. 2020; Zhang et al. 2020a). Particulate matter is one of the major pollutants in the NCP.

Particulate matter, especially PM$_{2.5}$ (particulate matter consisting of fine particles with aerodynamic diameters of less than 2.5 µm), plays important roles in the atmosphere, such as driving visibility reduction, acid deposition, and climate change (Cheng et al. 2021; Liu et al. 2019, 2021a; Nguyen et al. 2019; Zhang et al. 2020c; Zhao et al. 2020a). PM$_{2.5}$ is also associated with adverse health effects. Exposure to high concentrations of PM$_{2.5}$ has been found to result in increases in hospitalization and mortality rates (Honda et al. 2020; Lurie et al. 2019; Sugiyama et al. 2020; Zhang et al. 2020c). Lung diseases and other organic damage resulting from atmospheric heavy metals have been well-documented. It is noteworthy that the rate of premature deaths caused through PM$_{2.5}$ exposure in China is more than 1,300,000 people annually (Liu et al. 2016). Research to characterize PM$_{2.5}$ in the NCP has been conducted over the last two decades. Several studies have examined the general characteristics of chemical components in PM$_{2.5}$ and discussed their seasonal variations (Lim et al. 2020; Liu et al. 2021b; Wei et al. 2020; Yi et al. 2021; Zhao et al. 2020b; Shivani et al. 2019; Sharma et al. 2014, 2021; Pant and Harrison 2012). OC and SNA (SO$_4^{2-}$, NO$_3^-$, and NH$_4^+$) with high concentrations were found to be the main components of PM$_{2.5}$ in NCP. Investigations have focused on correlations between PM$_{2.5}$ components and the formation of secondary particles (Dong et al. 2020; Du et al. 2019; Wang et al. 2020; Yang et al. 2020). The evolutionary processes and formation mechanisms of heavy PM$_{2.5}$ pollution have been discussed (Guo et al. 2020a; Hu et al. 2021; Li et al. 2020a; Shi et al. 2020). Several studies have aimed to reveal the regional transportation and source apportionment of PM$_{2.5}$ (Feng et al. 2016; Wang et al. 2016; Yao et al. 2016; Zong et al. 2018). In addition, lidar, satellite remote sensing, and numerical simulation have been used to investigate PM$_{2.5}$ pollution (He et al. 2020; Li et al. 2020b; Lin et al. 2018; Lv et al. 2017).

Most previous studies have focused on the urban or background area of the NCP. The present study was conducted in Pingyin County, Jinan City, Shandong Province, which is located in the middle of the NCP and is a typical industrial area. Chemical characterization, source analysis, and a health risk assessment of PM$_{2.5}$ were conducted to provide reference data for policymakers to improve the air quality in the NCP.

Materials and methods

Sample collection

Manual sampling was conducted in the spring (20 May–11 June), summer (17 July–2 August), autumn (30 October–14 November), and winter (December) of 2019 at an air-quality monitoring station in Huaihai Yushu, Pingyin County, with samples collected over 15 days in spring, summer, and autumn, and 30 days in winter. The sampling periods included a total of 84 days, accounting for approximately one-quarter of the total days in the year. The ambient air particulate matter sampler instrument was produced by Dan-dong Baite Company and has been certified by the China National Environmental Monitoring Centre. One Teflon membrane and one quartz membrane (47-mm Whatman filters) were collected from two sets of instruments per sampling event. The sampling period was 23 h, with 1 h for membrane replacement, and a blank membrane was taken every 15 days. A total of 168 samples were obtained. The ratio of organic carbon (OC) to elemental carbon (EC), nine water-soluble ions, and 18 elements were analyzed for each membrane filter. The sampling location is shown in Fig. 1.

Analytical methods

Ion analysis

In this study, the water-soluble ions F$^-$, Cl$^-$, NO$_3^-$, SO$_4^{2-}$, Na$^+$, K$^+$, Mg$^{2+}$, Cu$^{2+}$, and NH$_4^+$ were analyzed with reference to the national standards HJ 799–2016 (2016) and HJ 800 (2016). A quarter of a quartz membrane filter was smashed, immersed in 10 ml of deionized water, shaken uniformly, and extracted for 15 min in an ultrasonic bath. The supernatant was removed after 5 min of settling and analyzed using Dionex ICS-2100 and ICS-1100 ion chromatographs.

OC/EC analysis

OC and EC were measured using a USA Sunset Lab RT-4 carbon analyzer. The samples were not pre-treated. After the quartz membrane was removed from the refrigerator and
allowed to come to room temperature, the analyzer was used to directly measure OC and EC.

**Element analysis**

In this study, 18 elements were analyzed, including Na, Mg, and Al. First, a Teflon membrane was placed into a clean sample digestion tank for digestion, then 20 ml of saturated boric acid solution was added for the complexation of excessive F⁻ ions, and closed microwave digestion was carried out. After digestion, the volume was adjusted to 50 ml.

Element analysis was conducted using an Intrepid-XDL spectrometer.

Before each chemical component measurement, standard samples from the Institute for Environmental Reference Materials of the Ministry of Environmental Protection were applied to calibrate the instruments. A multi-point calibration was done every week. Parallel samples contributed at least 10% of the total number of samples. In addition, SO₂ and NOx concentrations were monitored synchronously using 100E and 200E SO₂ analyzers produced by the API Corporation of America.

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**Figure 1** Location of the monitoring site. (A) The global PM$_{2.5}$ distribution from 2001 to 2006 based on satellite observations by the United States National Aeronautics and Space Administration (Van Donkelaar et al. 2010). (B) The national PM$_{2.5}$ distribution in China based on monitoring data from 1436 sites from 2014 to 2017 (Wang et al., 2019b). (C) Administrative map of Jinan City, with different colors indicating different counties. The black triangle represents the location of the monitoring site used in this study.
Source contribution analysis

The sources of PM$_{2.5}$ were analyzed using the positive matrix factorization (PMF) receptor model. First, the error associated with the chemical component weights of the receptor was determined, and then the main sources of contamination and their contribution ratios were determined using the least squares method. PMF is a type of multivariate factor analysis in which a mathematical method is used to decompose matrix X containing sample data for a species into two matrices: factor contributions (G) and factor spectra (F). The method does not require the input of a source spectrum and ensures that the decomposition factor contribution (G) and factor spectrum (F) are non-negative (Jain et al. 2020; Lv et al. 2021; Manousakas et al. 2017; Padoan et al. 2020). The matrix $X$ can be represented by the following formula:

$$x_{ij} = \sum_{k=1}^{p} g_{ik} f_{kj} + e_{ij}$$

where $x_{ij}$ is the concentration of species $j$ in sample $i$, $p$ is the number of factors, $g_{ik}$ is the contribution of factor $k$ to sample $i$, $f_{kj}$ is the concentration of factor $k$ in species $j$, and $e_{ij}$ is the error of species $j$ in sample $i$.

Health risk assessment

Heavy metal elements can enter the human body and endanger human health through three pathways: the skin, digestive system, and respiratory system. The heavy metal elements in PM$_{2.5}$ mainly enter the human body through the respiratory system. In this study, four heavy metals registered with the United States Agency for Toxic Substances and Disease Registry—As, Mn, Ni, and Pb—were selected from the monitored metals for analysis. The potential exposure risk was analyzed using the pollutant exposure model recommended by the United States Environmental Protection Agency (US EPA). The formula for calculating the exposure dose is as follows:

$$ADD(LADD) = C \times IR \times EF \times ED/(BW \times AT),$$

where ADD is the non-carcinogenic exposure dose with units of mg/(kg·day), LADD is the lifetime average exposure
dose of carcinogens (mg/[kg·day]), C is the concentration of heavy metals (mg/m$^3$), IR is the respiratory rate (m$^3$/day), EF is the exposure frequency (days/a), ED is the exposure duration (a), BW is body mass (kg), and AT is the average exposure time (day). The values of these parameters are shown in Table 1 (US EPA and Ministry of Ecology and Environment of the People’s Republic of China) (Bi et al. 2020; Li et al. 2021).

The carcinogenic risk index and non-carcinogenic risk index through respiratory exposure were calculated as follows:

$$R1 = ADD/RfD,$$

$$R2 = LADD \times SF,$$

where $R_1$ is the non-carcinogenic risk of a pollutant associated with respiratory exposure, $R_2$ is the carcinogenic risk of a pollutant associated with respiratory exposure, RfD is the reference dose (mg/[kg·day]), and SF is the carcinogenic intensity coefficient of a carcinogenic pollutant (kg·day/mg). When $R_1 \leq 1$, the non-carcinogenic health risk is negligible; when $R_1 > 1$, the pollutant poses a non-carcinogenic risk. When $R_2 < 10^{-6}$, the carcinogenic risk is negligible, when $R_2$ is between $10^{-6}$ and $10^{-4}$, there is a potential carcinogenic risk; when $R_2 > 10^{-4}$, there is a high carcinogenic risk. The values of RfD and SF are shown in Table 2 (US EPA and Ministry of Ecology and Environment of the People’s Republic of China) (Chen et al. 2015; Zhai et al. 2014).

| Heavy metal element | RfD (mg/[kg·day]) | SF (kg·day/mg) |
|---------------------|------------------|----------------|
| As                  | $3.0 \times 10^{-4}$ | 15.1           |
| Mn                  | $3.0 \times 10^{-4}$ | —              |
| Ni                  | $2.0 \times 10^{-2}$ | 1.19           |
| Pb                  | $3.5 \times 10^{-3}$ | 0.28           |

Table 1 Exposure parameters for heavy metals entering the human body through the respiratory system

| Population          | Age range (years) | IR (m$^3$/day) | EF (days/a) | ED (a) | BW (kg) | AT to non-carcinogenic materials (day) | AT to carcinogenic materials (day) |
|---------------------|-------------------|---------------|-------------|--------|---------|----------------------------------------|------------------------------------|
| Adult males         | 18–70             | 16.6          | 365         | 30     | 67.3    | $30 \times 365$                        | $70 \times 365$                    |
| Adult females       | 18–70             | 13.5          | 365         | 30     | 57.5    | $30 \times 365$                        | $70 \times 365$                    |
| Children and adolescents | 6–17             | 8.7           | 365         | 18     | 46      | $18 \times 365$                        | $70 \times 365$                    |
Results and discussion

PM$_{2.5}$ component concentrations

The concentrations of PM$_{2.5}$ components during the sampling period are shown in Fig. 2. The total concentration of 29 components was $53.4 \pm 43.9 \ \mu g\cdot m^{-3}$, with the largest contribution from the NO$_3^-$ ion, which had an average concentration of $14.6 \pm 14.2 \ \mu g\cdot m^{-3}$, followed by OC, SO$_4^{2-}$, and NH$_4^+$, with average concentrations of $9.3 \pm 5.5$, $9.1 \pm 6.4$, and $8.1 \pm 6.8 \ \mu g\cdot m^{-3}$, respectively. These four components were the main contributors to PM$_{2.5}$, and the sum of their concentrations accounted for 76.7% of the total concentration of 29 components. Among other components, the concentrations of EC and watersoluble ions such as Cl$^-$ and K$^+$ were between $0.2 \pm 0.1$ and $2.2 \pm 1.5 \ \mu g\cdot m^{-3}$, with Mg$^{2+}$ having the lowest concentration among components, and EC having the highest concentration. The concentrations of 18 elements including Na, Mg, and Al were between $2.3 \pm 1.6$ and $888.1 \pm 415.2 \ ng\cdot m^{-3}$, with Ni having the lowest concentration and K the highest.

Figure 3 shows the average variations in the concentrations of the main components of PM$_{2.5}$ in Pingyin County among seasons. The concentrations of OC, NO$_3^-$, and SO$_4^{2-}$ were highest in winter and lowest in summer. The mean concentration of OC was higher in winter ($13.8 \pm 5.7 \ \mu g\cdot m^{-3}$) than in autumn ($8.9 \pm 2.6 \ \mu g\cdot m^{-3}$), spring ($7.5 \pm 2.0 \ \mu g\cdot m^{-3}$), or summer ($3.5 \pm 2.3 \ \mu g\cdot m^{-3}$). OC is mainly derived from the combustion of fossil fuels, and its concentration is elevated in winter due to emissions from the burning of coal for heating. High concentrations of OC in autumn and spring are generally related to biomass combustion (Ikemori et al. 2021; Khan et al. 2021; Ren et al. 2021). The mean concentration of NO$_3^-$ was higher in winter ($23.4 \pm 15.0 \ \mu g\cdot m^{-3}$) than in autumn ($19.9 \pm 13.5 \ \mu g\cdot m^{-3}$), spring ($5.1 \pm 3.1 \ \mu g\cdot m^{-3}$), or summer ($3.1 \pm 3.0 \ \mu g\cdot m^{-3}$). NO$_3^-$ mainly originates from the emissions of motor vehicle exhaust and industrial processes. The concentrations of NO$_3^-$ were significantly higher in autumn and winter than in spring and summer in Pingyin County, indicating that industrial emissions have a strong influence on NO$_3^-$. 

![Fig. 2. Average concentrations of the chemical components of PM$_{2.5}$ during the sampling period.](image-url)
The average concentration of $\text{SO}_4^{2-}$ was greater in winter ($12.4 \pm 7.6 \, \mu\text{g} \cdot \text{m}^{-3}$) than in autumn ($8.2 \pm 4.0 \, \mu\text{g} \cdot \text{m}^{-3}$), spring ($7.1 \pm 4.3 \, \mu\text{g} \cdot \text{m}^{-3}$), or summer ($5.7 \pm 4.7 \, \mu\text{g} \cdot \text{m}^{-3}$). $\text{SO}_4^{2-}$ mainly arises through the conversion of $\text{SO}_2$ from coal-fired emissions. In contrast with OC and $\text{NO}_3^-$, concentrations of $\text{SO}_4^{2-}$ were similar in summer, spring, and autumn. Although $\text{SO}_2$ emissions are low in summer, the high temperatures and humidity are favorable for the generation of $\text{SO}_4^{2-}$ (Wang et al. 2019a; Yang et al. 2018). $\text{NH}_4^+$ exhibited a different seasonal pattern, with higher concentrations in summer than in spring. The mean concentration of $\text{NH}_4^+$ was higher in winter ($12.1 \pm 6.7 \, \mu\text{g} \cdot \text{m}^{-3}$) than in autumn ($9.8 \pm 5.7 \, \mu\text{g} \cdot \text{m}^{-3}$), summer ($3.2 \pm 1.8 \, \mu\text{g} \cdot \text{m}^{-3}$), or spring ($2.2 \pm 1.6 \, \mu\text{g} \cdot \text{m}^{-3}$). $\text{NH}_4^+$ mainly originates from coal combustion, agricultural production, and organic matter degradation. The high concentration of $\text{NH}_4^+$ in summer may be related to the high temperatures, which increases the volatility of $\text{NH}_3$ and its rate of chemical conversion (Pandolfi et al. 2012). For typical heavy metals, overall concentrations were higher in autumn and winter, and lower in spring and summer. The concentrations of Mn and Pb were similar, with no seasonal variation apparent. The concentration of Ni was similar in spring, summer, and autumn, and higher in winter. The concentration of As was highest in autumn, lowest in summer, and similar in winter and spring.

### Sulfur oxidation rate and nitrogen oxidation rate

The sulfur oxidation rate (SOR) and nitrogen oxidation rate (NOR) are used to indicate the degrees of conversion of $\text{SO}_2$ and $\text{NO}_2$ in the atmosphere, respectively, and are defined as $\text{SOR} = \rho(\text{SO}_4^{2-})/(\rho(\text{SO}_4^{2-}) + \rho(\text{SO}_2))$ and $\text{NOR} = \rho(\text{NO}_3^-)/(\rho(\text{NO}_3^-) + \rho(\text{NO}_2))$. Higher SOR and NOR values indicate greater oxidation of gaseous pollutants and the generation of more $\text{SO}_4^{2-}$ and $\text{NO}_3^-$ (Ding et al. 2019; Guo et al. 2020b).

Fig. 4 shows SORs and NORs in different seasons in Pingyin County. Throughout the year, the mean SOR and NOR were $0.30 \pm 0.14$ and $0.21 \pm 0.12$, respectively. The $\text{NO}_3^-$ concentration in Pingyin County was higher than the $\text{SO}_4^{2-}$ concentration, whereas sulfur underwent oxidation at a higher rate than did nitrogen. In autumn, the NOR was greater than the SOR, but it was lower than the SOR in spring, summer, and winter. Seasonal variations in the SOR and NOR were observed. The SOR was higher in winter ($0.35 \pm 0.14$) than in summer ($0.31 \pm 0.18$), spring ($0.27 \pm 0.11$), or autumn ($0.25 \pm 0.09$), and the highest concentration of $\text{SO}_4^{2-}$ was observed in winter. Although $\text{SO}_2$ emissions are low in summer, the conversion rate is high. The SOR was similar in spring and autumn. The NOR was higher in winter ($0.27 \pm 0.11$) than in autumn ($0.27 \pm 0.12$), spring ($0.15 \pm 0.06$), or summer ($0.11 \pm 0.09$), with similar levels in winter and autumn that were higher than the values in spring and summer. The high NOR in autumn is likely one
driver of the high NO$_3^-$ concentration in autumn relative to spring and summer.

**Secondary OC estimation**

The OC/EC ratio is often used to characterize the emission and transformation of carbon aerosols and to evaluate and identify secondary sources of particulate matter. EC, which mainly arises from incomplete combustion of carbonaceous fuel, is stable and does not participate in chemical reactions in the atmosphere. Therefore, it is used as a tracer for primary anthropogenic emissions. OC includes primary OC (POC), which is directly discharged from emission sources, as well as secondary OC (SOC), which is generated by photochemical reactions. Chow et al. (1996) argued that an OC/EC ratio > 2 indicates conditions under which SOC can be generated in the atmosphere, whereas Castro et al. (1999) proposed that SOC can be generated when the OC/EC ratio
is > 1.1. In addition to photochemical reactions, biomass burning releases a large amount of OC and its effect on EC is relatively small, leading to a high OC/EC ratio. Zhang et al. (2007) found that the average OC/EC in China reached 7.7 when grain straw was burned.

As the complicated atmospheric formation processes and condensation/distribution mechanisms of SOC remain unclear, no unified analytical method is available for the direct measurement of SOC. In addition to directly simulating SOC generation under specific conditions in an aerosol chamber, SOC concentrations in the atmospheric environment are generally estimated through indirect methods, such as calculation from the OC/EC concentration ratio, the organic molecular tracing method, and the numerical model prediction method. Among these methods, the one based on the OC/EC ratio is the simplest and most direct and has been widely applied to identify and assess SOC pollution.

In the OC/EC-ratio method, the OC/EC concentration ratio of particulates emitted from pollution sources is considered to be relatively stable and associated with the emission source. When the OC/EC ratio of atmospheric particles exceeds this characteristic stable value, SOC formation is likely. According to this theory, Turpin and Huntzicker (1995) proposed the following formula for calculating SOC:

\[
SOC = TOC - EC \times (OC/EC)_{pri}
\]

where TOC represents total OC, and \((OC/EC)_{pri}\) represents the average OC/EC ratio in the pollution source, which is difficult to determine and associated with high uncertainty. Estimating the ratio requires understanding the emission characteristics of each pollution source in the region and the consideration of daily and seasonal fluctuations in both emissions and meteorological conditions. Due to this difficulty, Castro et al. (1999) proposed the following formula to estimate SOC based on minimum OC/EC values:

\[
SOC = TOC - EC \times (OC/EC)_{min}
\]

The OC/EC ratios in different seasons in Pingyin County, the SOC concentrations estimated using this method, and SOC/OC ratios are shown in Fig. 5. These results revealed an annual average OC/EC ratio of 4.7 ± 1.1, indicating the presence of SOC generation. The OC/EC ratio was higher in spring (5.4 ± 0.7) than in autumn (5.3 ± 0.6), winter (4.4 ± 0.8), or summer (3.8 ± 2.0), with the higher values in spring and autumn indicating biomass combustion. The average SOC concentration over Pingyin County was 2.8 ± 1.9 μg·m⁻³ throughout the year, and the seasonal SOC concentrations were 4.0 ± 2.0 μg·m⁻³ in winter, 3.3 ± 2.0 μg·m⁻³ in summer, 1.7 ± 0.7 μg·m⁻³ in spring, and 1.6 ± 0.9 μg·m⁻³ in autumn. Although the OC concentration was low in summer, the SOC concentration was high then. The annual average SOC/OC ratio was 0.3 ± 0.2, which indicates that approximately 30% of the OC concentration in Pingyin County was SOC generated through secondary chemical reactions, whereas the proportion of POC was approximately 70%. The SOC/OC ratio was 0.6 ± 0.3 in summer, 0.3 ± 0.1 in winter, 0.2 ± 0.1 in spring, and 0.2 ± 0.1 in autumn. Thus, the SOC/OC ratio was highest in summer, and lowest in spring and autumn.

**Health risk assessment of heavy metals**

The carcinogenic and non-carcinogenic risk coefficients for four heavy metals were calculated, and the results are shown in Fig. 6. The non-carcinogenic risk coefficients for all heavy metals were lower than 1, indicating that the non-carcinogenic risk was negligible. No risk of cancer was associated with Mn, and the carcinogenic risk coefficient for Ni was less than 10⁻⁶, indicating negligible risk. The risk coefficient associated with Pb exposure in children and adolescents was...
less than $10^{-6}$, indicating negligible risk, but the carcinogenic risk coefficient for adults was between $10^{-6}$ and $10^{-4}$, indicating a potential risk due to Pb. The carcinogenic risk coefficient associated with As exposure in adults and children and adolescents was between $10^{-6}$ and $10^{-4}$, indicating that both groups face potential risk. From the comparison among groups, the carcinogenic and non-carcinogenic risk coefficients associated with heavy metals were greatest for adult males, followed by adult females, and were lowest for children and adolescents.

**PM$_{2.5}$ source contribution analysis**

The input species were classified into strong, weak, and bad based on the signal-to-noise ratio (S/N). Generally, species with S/N > 2.0 were categorized as strong; those with 0.2 < S/N < 2.0 were categorized as weak; and those with S/N < 0.2 were categorized as bad and were excluded from the PMF analysis. PMF factor numbers of 3–8 were explored to determine the optimum solution, and the 5-factor result was considered the most reasonable since it minimized the objective residual function.

PM$_{2.5}$ sources were classified into coal, traffic, dust, industry, and other sources based on the PM$_{2.5}$ source analysis results published by the Jinan municipal government. Fig 7 shows the spectra of various sources based on the optimal solution of the PMF model. For factor 1, the contributions of secondary components such as SO$_4^{2-}$, NO$_3^-$, and NH$_4^+$ were high; therefore, this factor was considered the secondary source factor. For factor 2, the contributions of Na, S, P, and other elements were high, which is characteristic of chemical production emissions (Wang et al. 2021); therefore, this factor was considered to represent industrial sources. For factor 3, the contribution of the element Zn was the highest, indicating traffic emissions (Hao et al. 2019), and the high contents of OC and EC also support its designation as the traffic source factor. For factor 4, the contributions of crustal elements such as Al, Si, Ca, and Fe were high, so it was identified as the dust source factor. For factor 5, Cl$^-$ made the greatest contribution, which is characteristic of coal emissions (Sawlani et al. 2021), and the OC concentration was high, indicating that this factor represents coal combustion.

Figure 8a shows the contributions of various PM$_{2.5}$ sources in Pingyin County based on the PMF results. The largest contribution was from secondary sources, which accounted for 45.4%, followed by coal-fired sources at 24.9%, traffic sources at 17.5%, and dust and industrial sources, which accounted for 8.4% and 3.8%, respectively. From the PMF analytical results, secondary sources were attributed to primary sources based on an atmospheric pollution source emission inventory for Pingyin County, and the final results are shown in Fig. 8b. Coal-fired sources contributed the most to PM$_{2.5}$ in the region, accounting for 35.9% of the total; traffic sources contributed a similar amount as coal-fired sources, at 32.1%. The third-largest contributor was industrial sources, which accounted for 17.2%. The contributions of dust and other emission sources to PM$_{2.5}$ were 8.4% and 6.4%, respectively. Thus, emissions from coal and traffic sources contributed the most to PM$_{2.5}$, and therefore, the regulation of coal burning and traffic emissions should be strengthened to improve air quality in Pingyin County. In this study, coal-fired sources include coal-fired industrial activities, whereas industrial sources refer to non-coal-fired industries.

Coal-burning and traffic emissions were the largest contributors to PM$_{2.5}$ in this region. Therefore, policymakers should take steps to control coal-burning and traffic emissions to reduce the concentration of PM$_{2.5}$ in typical industrial areas in the NCP, such as adopting measures to...
increase the proportion of clean energy, eliminate inefficient coal-fired boilers, promote renewable-energy vehicles, and upgrade the emission standards for motor vehicles.

**Conclusion**

In 2019, the total concentration of 29 components of PM$_{2.5}$ over Pingyin County was $53.4 \pm 43.9$ μg·m$^{-3}$, with the largest contribution from NO$_3^-$ ($14.6 \pm 14.2$ μg·m$^{-3}$), followed by OC, SO$_4^{2-}$, and
NH₄⁺, which had average concentrations of 9.3 ± 5.5, 9.1 ± 6.4, and 8.1 ± 6.8 µg·m⁻³, respectively. The SOR and NOR were 0.30 ± 0.14 and 0.21 ± 0.12, respectively, in 2019. Aside from autumn, when the NOR was greater than the SOR, the SOR was higher than the NOR in all other seasons. The mean NO₃⁻ concentration over Pingyin County was higher than the SO₄²⁻ concentration, whereas the mean NOR was lower than the mean SOR.

The annual average concentration of SOC in 2019 was estimated to be 2.8 ± 1.9 µg·m⁻³, and SOC concentrations were 4.0 ± 2.0, 3.3 ± 2.0, 1.7 ± 0.7, and 1.6 ± 0.9 µg·m⁻³ in winter, summer, spring, and autumn, respectively. The health risk assessment of typical heavy metal elements showed that Pb poses a potential carcinogenic risk for adults, and As poses potential carcinogenic risks for adults, children, and adolescents. The results of PMF analysis showed that coal-burning sources contributed the largest fraction of total PM₂.₅, at 35.9%, whereas the contribution from traffic sources was similar, at 32.1%. The third-largest contributor to PM₂.₅ was industrial sources, which accounted for 17.2% of the total. The contributions of dust and other emission sources to PM₂.₅ were 8.4% and 6.4%, respectively.

Author contribution Zhanshan Wang: conceptualization; methodology; formal analysis; investigation; writing—original draft; visualization.
Zhiheng Li and Chen Guo: conceptualization, validation, investigation, writing—review and editing.
Kai Wu and Puzhen Zhang: software, investigation, resources.
Xiaoqian Li and Jiayi Yan: conceptualization; methodology; validation; writing—review and editing; supervision.
Zhaoxin Sun and Xiaojing Zhu: conceptualization, validation, resources.
Yongjie Wei: software, investigation, resources, project administration.

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Data availability The data are available on request to the lead corresponding author.

Declarations

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